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Sandia National Laboratories
Waste Isolation Pilot Plant

Responses to Three EPA Comments Pertaining to Comparisons of Measured and Predicted Dissolved and Colloidal Th(IV) and Am(III) Concentrations

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1 INTRODUCTION

This analysis responds to three closely related U.S. Environmental Protection Agency (EPA) comments arising from its completeness review of the U.S Department of Energy's (DOE's) second Compliance Recertification Application (CRA-2009) (U.S. DOE, 2009) for the Waste Isolation Pilot Plant (WIPP). These comments all pertain to comparisons of measured and predicted dissolved and/or colloidal Th(IV) and Am(III) concentrations. They are: (1) Comment 8 of the EPA's January 25, 2010, follow-up comments to the issues first raised in Comment 1-23-6 of its May 21, 2009 letter to the DOE (Cotsworth, 2009); (2) Comment 4-C-35 of the EPA's February 22, 2010, letter to the DOE (Kelly, 2010); and (3) Comment 4-C-36 of the EPA's February 22, 2010 letter to the DOE. Subsections 3.1, 3.2, and 3.3 reproduce these comments in their entirety.

Comment 8 of the EPA's January 25, 2010, follow-up comments to the issues first raised in Comment 1-23-6 of its May 21, 2009 letter to the DOE (Cotsworth, 2009) stated that:

> DOE should perform FMT calculations to predict the concentrations of Nd(III) under the conditions of the Nd(III) solubility experiments in the NaCl, GWB, and ERDA-6 brines. These calculations would provide direct evidence of the extent of the differences between the experimental results and the FMT calculated solubilities, which will be useful even though experimental data are unavailable at higher pcH in GWB. For the carbonate experiments, it may be useful to perform calculations using $Am(OH)_3(s)$ as the solubility-controlling solid as well as calculations using $AmOHCO_3(s)$ as the solubility controlling solid, to indicate potential solution composition changes that might be caused by a change in the stable phase or mixed solid phase formation.

Comment 4-C-35 of the EPA's February 22, 2010, letter to the DOE (Kelly, 2010) stated that:

> A review of the recent literature regarding aqueous thorium speciation in carbonate solutions indicates that the speciation selected by the OECD critical review (Rand et al., 2009) is the most consistent with the available data. The thorium-carbonate and –hydroxycarbonate speciation and stability constants in the FMT database differ from the Rand et al. (2009) review and compilation and should be revised. However, such a revision of the FMT database will involve a significant effort because Pitzer parameters are unavailable for the thorium-hydroxycarbonate species identified by Rand et al. (2009) as the most likely significant species, including $ThOH(CO_3)_4^{5-}$, $Th(OH)_2(CO_3)_2^{2-}$, and $Th(OH)_4CO_3^{2-}$. Because of the low CO_2 fugacities established by the brucite-hydromagnesite buffer in the WIPP repository, it is uncertain whether

replacing the current thorium-hydroxycarbonate and -carbonate speciation and stability constants in FMT with the revised thorium-hydroxycarbonate and -carbonate species from Rand et al. (2009) would significantly affect calculated thorium concentrations under WIPP repository conditions. In DOE's response to Completeness Comment 1-23-3, Borkowski and Richmann (2009) indicated that the FMT-calculated thorium concentrations remain conservative. However, no quantitative evaluation was presented to support this assertion.

As part of their evaluation of actinide solubility uncertainties for the CRA-2009 PABC, DOE has presumably used FMT to calculate expected thorium concentration in carbonate-bearing solutions in the experiments reported by Östhols et al. (1994), Rai et al. (1995), Felmy et al. (1997), Altmaier et al. (2005), and Altmaier et al. (2006). DOE should examine whether the thorium concentrations predicted by FMT modeling consistently differ from the reported experimentally measured thorium concentrations in carbonate-bearing solutions. Realistic or conservative predicted +IV actinide solubilities would be indicated if the FMT predicted concentrations are consistently the same or higher than the concentrations measured in the experiments with carbonate. For the CRA-2014 PA, DOE should evaluate the available data in an effort to derive Pitzer parameters for the thorium-hydroxycarbonate species $ThOH(CO_3)_4^{5-}$, $Th(OH)_2(CO_3)_2^{2-}$, and $Th(OH)_4CO_3^{2-}$ and update the thorium aqueous speciation data in the FMT database.

Comment 4-C-36 of the EPA's February 22, 2010 letter to the DOE (Kelly, 2010) stated:

An experimental investigation reported by Altmaier et al. (2004) has indicated that intrinsic thorium colloids (eigencolloids) can form and remain stable at high ionic strength (up to 5 M NaCl or 4.5 M $MgCl_2$). The resulting total mobilized thorium concentrations (dissolved plus intrinsic colloids) appear to be independent of ionic strength, with a mean $\log[Th]_{total} \approx \log[Th]_{colloidal} = -6.3 \pm 0.5$. This reported intrinsic colloid concentration exceeds the PABC 2009-calculated dissolved thorium concentration of $\log[Th]_{dissolved} = -7.2$ by approximately an order of magnitude. The colloidal Th concentration reported by Altmaier et al. (2004) is not accounted for in the implementation of the colloidal actinide source term model in PA because intrinsic Th(IV) colloids were assumed not to be present, based on an evaluation of the literature for the Compliance Certification Application and the colloidal actinide source term conceptual model peer review.

Altmaier et al. (2004) also observed the formation of colloidal $Mg_2(OH)_3Cl{\cdot}4H_2O$ that sorbed thorium (producing thorium pseudocolloids or mineral fragment colloids). In solutions saturated with respect to $Mg_2(OH)_3Cl{\cdot}4H_2O$, the thorium concentration was reported to be $\log[Th]_{total} = -4.8 \pm 0.2$. In comparison, the mineral fragment colloid concentration used in PA for thorium(IV) is much lower, with $\log[Th]_{mineral\ fragment\ colloids} = -7.6$. Altmaier et al. (2004) noted that at lower magnesium concentrations, $Mg_2(OH)_3Cl{\cdot}4H_2O$(cr)

is not stable and these colloids would not form. However PABC-2009 calculations with GWB brine indicate that $Mg_2(OH)_3Cl{\cdot}4H_2O$(cr) is stable under WIPP repository conditions, so that formation of these pseudocolloids in Salado brines cannot be ruled out on this basis. Altmaier et al. (2004) stated that the relatively small solid to solution ratios in their experiments are not applicable to a repository environment where sorption of thorium on the large amounts of $Mg_2(OH)_3Cl{\cdot}4H_2O$(cr) would prevail over sorption onto the mobile colloids. However, it is not clear that such an argument is consistent with the existing WIPP colloidal actinide source term conceptual model.

DOE should address whether significant thorium intrinsic colloids and pseudocolloids could form in the WIPP repository. Unless the formation of such colloids can be ruled out by the available data, DOE should address the possible effects of such colloid formation on repository performance.

The analysis described in this report was carried out under Subsection 2.3, "Other Analysis or Information Requests," of Nuclear Waste Management Procedure 9-1, Rev. 8, "Analyses."

Table 1 (see next page) defines the abbreviations, acronyms, initialisms, etc., used in this analysis report.

Table 1. Abbreviations, Acronyms, Initialisms, etc.

Abbreviation, Acronym, or Initialism	Definition
Am, Am(III)	americium, americium in the +III oxidation state
am	amorphous
anhydrite	$CaSO_4$
brucite	$Mg(OH)_2$
C	carbon
Cl, Cl^-	chloride, chloride ion
CO_3, CO_3^{2-}	carbonate, carbonate ion
CMS	(Sandia/WIPP software) Configuration Management System
CPG	(SNL's) Carlsbad Programs Group
cr	crystalline
CRA-2004	the first WIPP Compliance Recertification Application, submitted to the EPA in March 2004
CRA-2009	the second WIPP Compliance Recertification Application, submitted to the EPA in March 2009
DBR	direct brine release
DOE	(U.S.) Department of Energy
EPA	(U.S.) Environmental Protection Agency
ERDA-6	Energy Research and Development Administration (WIPP Well) 6, a synthetic brine representative of fluids in Castile brine reservoirs
Fm.	Formation
FMT	Fracture-Matrix Transport, a geochemical speciation and solubility code
GWB	Generic Weep Brine, a synthetic brine representative of intergranular Salado brines
halite	NaCl
HCO_3, HCO_3^-	bicarbonate, bicarbonate ion
hydromagnesite	$Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$
I	ionic strength
kD	kilo Daltons
log	the common logarithm or logarithm (base 10)
M	molar
m	molal
$MgCl_2$	magnesium chloride
MgO	magnesium oxide, used to refer to the WIPP engineered barrier, which includes periclase as the primary constituent and various concentrations of impurities

Table 1 continued on next page

Table 1. Abbreviations, Acronyms, Initialisms, etc. (continued).

Abbreviation, Acronym, or Initialism	Definition
Na, Na^+	sodium, sodium ion
Nd, Nd(III)	neodymium, neodymium in the +III oxidation state
nm	nanometer(s)
NP	(SNL/WIPP) Nuclear Waste Management Procedure
OH, OH^-	hydroxide, hydroxide ion
Np, Np(IV)	neptunium, neptunium in the +IV oxidation state
PA	performance assessment
PABC	(WIPP) Performance Assessment Baseline Calculations, carried out in 2005 and 2009
PAVT	(WIPP) Performance Assessment Verification Test, carried out in 1997
pcH	the negative log (base 10) of the molar concentration of H^+
periclase	pure, crystalline MgO, the primary constituent of the WIPP engineered barrier
phase 3	$Mg_2(OH)_3Cl{\cdot}4H_2O$
phase 5	$Mg_3(OH)_5Cl{\cdot}4H_2O$
Pu, Pu(IV)	plutonium, plutonium in the +IV oxidation state
Rev.	Revision
SNL	Sandia National Laboratories
Th, Th(IV)	thorium, thorium in the +IV oxidation state
TIC	total inorganic C (the sum of the dissolved species of inorganic C)
U, U(IV)	uranium, uranium in the +IV oxidation state
WIPP	(U.S. DOE) Waste Isolation Pilot PIant

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2 METHODS

This section presents the methods we used for our responses to three closely related EPA comments arising from its completeness review of the CRA-2009 (U.S. DOE, 2009) for the WIPP.

2.1 Comment 8 of the Follow-Up Comments to 1-23-6

Comment 8 of the EPA's January 25, 2010, follow-up comments to the issues first raised in Comment 1-23-6 of its May 21, 2009 letter to the DOE (Cotsworth, 2009) requested that we "perform FMT calculations to predict the concentrations of Nd(III) under the conditions of the Nd(III) solubility experiments in the NaCl, GWB, and ERDA-6 brines."

Therefore, we compared all of the Nd(III) solubilities measured by Borkowski et al. (2009) with those predicted by FMT (Babb and Novak, 1997 and addenda; Wang, 1998) for the numerical values of the conditions reported by Borkowski (2010).

Borkowski (2010) provided the numerical values of the final, measured values of the pcH (the negative log (base 10) of the molar concentration of H^+), the initial carbonate (CO_3^{2-}) concentrations, and the final, measured Nd(III) concentrations from all 237 of his Nd(III) solubility experiments (85 in 5 M NaCl, 78 in ERDA-6, and 74 in GWB). Energy Research and Development Administration (WIPP Well) 6 (ERDA-6) and Generic Weep Brine (GWB) are two standard WIPP brines that are frequently used for laboratory and modeling studies that support WIPP performance assessment (PA). ERDA-6 is a synthetic brine representative of fluids in brine reservoirs in the Castile Fm. (Popielak et al., 1983). Snider (2003) verified that GWB is the average composition of intergranular fluids collected from the Salado Formation (Fm.) at the original stratigraphic horizon of the repository and analyzed by Krumhansl et al. (1991).

Borkowski (2010) provided the numerical values of these parameters because Borkowski et al. (2009) included only scatter plots, and because using the numerical values provided by Borkowski (2010) was more accurate than using an application such as Data Thief to estimate numerical values from the figures in Borkowski et al. (2009). Borkowski (2010) also specified whether each experiment was an oversaturation or an undersaturation experiment.

We used the final, measured values of the pcH directly in our FMT input files. We assumed that the initial CO_3^{2-} concentrations reported by Borkowski (2010) corresponded to the initial total inorganic carbon (TIC) concentrations, and used these initial TIC concentrations in our input files. In other words, we assumed that the CO_3^{2-} initially present either continued to speciate essentially entirely as CO_3^{2-}, or converted to some mixture of CO_3^{2-} and HCO_3^-,

depending on the pcH of each experiment. We also assumed that the final, measured Nd(III) concentrations from these experiments corresponded to the Nd(III) solubilities for the conditions reported by Borkowski (2010). We modeled the oversaturation and the undersaturation experiments identically.

We used the initial TIC concentrations in our FMT input files along with the final values of pcH and the final Nd(III) solubilites because Borkowski et al. (2009) did not measure the final CO_3^{2-} concentration or the final TIC concentration in most cases. In a few cases, Borkowski measured the final TIC concentrations and concluded that these concentrations decreased to approximately 50–80% of their initial values, probably because of precipitation of unidentified CO_3^{2-}–bearing solids during the experiments. We cannot provide a reference for these posttest TIC analyses, the results of which were provided during a telephone conversation between Borkowski and Brush on February 12, 2010.

Subsection 3.1 (see below) provides the final, measured values of the pcH, the initial CO_3^{2-} concentrations, the run type, and the final, measured Nd(III) concentrations (solubilities) provided by Borkowski (2010, Tables 2–8).

In this analysis, Y.-L. Xiong used the final measured values of pcH, the initial CO_3^{2-} concentrations, and the composition of each solution to set up FMT input files to predict the solubilities of Nd(III) for each of the experiments summarized in Borkowski (2010). For GWB and ERDA-6, Xiong used the compositions given in Table 1 of Borkowski (2010). Xiong used these compositions instead of those specified by Popielak et al. (1983) for ERDA-6 and Snider (2003) for GWB because Borkowski et al (2009) used ERDA-6 and GWB diluted to 95% of the specified concentrations to avoid possible precipitation of evaporite minerals and concomitant precipitation of Nd(III) during his experiments. If Borkowski (2010) reported identical conditions for two or more experiments (i.e., identical values of the final pcH and initial CO_3^{2-} concentration), Xiong carried out only one FMT calculation for these experiments. Subsection 2.4 (below) provides information on the version of FMT and its supporting thermodynamic database that were used for these calculations, and other run-control information.

2.2 Comment 4-C-35

Comment 4-C-35 of the EPA's February 22, 2010, letter to the DOE (Kelly, 2010) requested that "[the] DOE should examine whether the thorium concentrations predicted by FMT modeling consistently differ from the ... experimentally measured [Th] concentrations in carbonate-bearing solutions" reported by "Östhols et al. (1994), Rai et al. (1995), Felmy et al. (1997), Altmaier et al. (2005), and Altmaier et al. (2006)."

We did not include any of the results from Östhols et al. (1994) in this analysis because: (1) all of their solutions had ionic strengths (I's) less than 3 M, and (2) Xiong et al. (2005) had observed that FMT significantly overpredicted the Th(IV) solubilities measured by Felmy et al. (1991) in 0.6 M NaCl, 1.2 M NaCl, and 0.6 M KCl solutions, and excluded these

and other results from solutions with I < 3 M. Xiong et al. (2009, p. 14, criterion S1) also excluded all of the results of Östhols et al. (1994) from their Th(IV) uncertainty analysis.

We included seven results from Rai et al. (1995, Figure 4) in this analysis because the experiments that yielded these results were carried out with Th(IV) and because the solutions that they used had I ≥ 3 M or m. We also included 19 results from Rai et al. (1995, Figure 5) in this analysis for the same reasons. We excluded all of the other results of Rai et al. (1995) because the runs that yielded them were conducted with U(IV) and/or with solutions with I < 3 M or m, and thus failed to satisfy criteria G6 and/or S1 of Xiong et al. (2009, p. 14).

For the results from Rai et al. (1995) that we included in this analysis, Xiong used the commercially available software application Data Thief to obtain numerical values of the logs of the CO_3^{2-} and Th concentrations from their data points plotted in Figure 4, and numerical values of the logs of the NaOH and Th concentrations from their data in Figure 5. Xiong then used FMT to predict the solubilities of Th(IV) under these conditions (see Subsection 2.4 below for the version of FMT and the thermodynamic database used for these calculations, and for other run-control information).

We did not reuse any of the data from Felmy et al. (1997) in this analysis because their paper is a review paper that included only previously published data that had already been included or excluded according to the criteria of Xiong et al. (2009) and/or this analysis. For example, both Xiong et al. (2009) and the authors of this analysis excluded all of the data of Östhols et al. (1994); and both Xiong et al. (2009) and this analysis included some data but excluded other data from Rai et al. (1995) (see the discussions of these two papers above).

Finally, in response to this comment, we included comparisons of 4 Th(IV) solubilities measured by Altmaier et al. (2005) and 12 Th(IV) solubilities measured by Altmaier et al. (2006) with those predicted by FMT. Subsection 2.4 provides information on the version of FMT and the thermodynamic database used for these calculations, and other run-control information.

2.3 Comment 4-C-36

EPA Comment 4-C-36 requests that, in view of the results reported by Altmaier et al. (2004), the DOE address (1) whether significant concentrations of Th(IV) intrinsic colloids (eigencolloids) or mineral-fragment colloids (pseudocolloids) could form in the WIPP, and (2) if so, what would be the effects of such colloids on PA. Subsection 3.3.1 describes the results of Altmaier et al. (2004) that pertain to this EPA comment.

Subsection 3.3 (see below) explains why the types and concentrations of colloids reported by Altmaier et al. (2004) do not appear to be relevant to the WIPP. Because we concluded that these colloids will not form in the WIPP, we did not attempt to assess the effects of these colloids on PA.

2.4 Software and Run Control

Xiong used the thermodynamic speciation and solubility code FMT (Babb and Nowak, 1997 and addenda), Version 2.4 (Wang, 1998), and the thermodynamic database FMT_050405.CHEMDAT (Nowak, 2005; Xiong, 2005) to predict Nd(III) solubilities for the comparisons with the experimentally measured values of Borkowski et al. (2009), Borkowski (2010), Rai et al. (1995), Altmaier et al. (2005), and Altmaier et al. (2005). This is the same database used by Brush and Xiong (2005), Brush (2005), and Xiong et al. (2005) for the baseline actinide solubilities and the solubility uncertainty ranges and probability distributions for the first WIPP Compliance Recertification Application (CRA-2004) Performance Assessment Baseline Calculation (PABC) and the CRA-2009 PA; and by Brush and Xiong (2009a, 2009c) and Xiong et al. (2009) for the baseline solubilities and the solubility ranges and distributions for the CRA-2009 PABC. Table 2 provides additional details on the software used for this analysis.

Table 2. Software Used for This Analysis.

Code	Version	Executable	Build Date	CMS Library	CMS Class
FMT	2.4	FMT_QB0204.EXE	09-03-98	LIBFMT	QB0204

J. J. Long carried out the FMT calculations under the PA run-control system used for WIPP compliance-related calculations. Tables 3 and 4 provide run-control information for these calculations. The versions of the FMT code and database used for this analysis are stored in the Sandia/WIPP Configuration Management System (CMS) libraries. Typing "libfmt" accesses the FMT library. The code and database are stored in the directory PACMS: [CMS_WIPP_NONPA.FMT]. All of the calculations used for this analysis, except those for Rai et al. (1995), are in class LANL09-0 of the CMS library LIBLANL09_FMT. The calculations for Rai et al. (1995) are in class PABC09-0 of LIBPABC09_FMT.

Table 3. Run-Control Information for All of the FMT Calculations Carried Out for This Analysis Except for Those Carried Out for the Comparisons with Rai et al. (1995).

	File Names[1,2,3]	CMS Library	CMS Class
Script	EVAL_FMT.COM	LIBLANL09_EVAL	LANL09-0
Script Input	EVAL_FMT_LANL09_*t*.INP	LIBLANL09_EVAL	LANL09-0
Script Log	EVAL_FMT_LANL09_*t*.LOG	LIBLANL09_FMT	LANL09-0
FMT:			
Input	FMT_050405.CHEMDAT	LIBLANL09_FMT	LANL09-0
Input	FMT_GENERIC.RHOMIN	LIBLANL09_FMT	LANL09-0
Input	FMT_LANL09_*w*_*n*.IN	LIBLANL09_FMT	LANL09-0
Input	FMT_LANL09_ *w*_*n*.INGUESS	LIBLANL09_FMT	LANL09-0
Output	FMT_LANL09_ *w*_*n*.OUT	LIBLANL09_FMT	LANL09-0
Output	FMT_LANL09_ *w*_*n*.FOR088	Not kept	Not kept

Footnotes for Table 3 provided on next page

Footnotes for Table 3:

1.

$t \in \{$ ERDA6_001M_C_PCH, ERDA6_01M_C_PCH, ERDA6_1E-4M_C_PCH, ERDA6_1E-5M_C_PCH, ERDA6_PCH, GWB_1E-2M_C_PCH, GWB_1E-3M_C_PCH, GWB_1E-4M_C_PCH, GWB_1E-5M_C_PCH, GWB_PCH, NACL_5M_1E-2M_C_PCH, NACL_5M_1E-3M_C_PCH, NACL_5M_1E-4M_C_PCH, NACL_5M_1E-5M_C_PCH, NACL_PCH $\}$

2.

$w \in \{$ ERDA-6_001M_C_PCH, ERDA-6_01M_C_PCH, ERDA-6_1E-4M_C_PCH, ERDA-6_1E-5M_C_PCH, ERDA-6_PCH, GWB_1E-2M_C_PCH, GWB_1E-3M_C_PCH, GWB_1E-4M_C_PCH, GWB_1E-5M_C_PCH, GWB_PCH, NACL_5M_1E-2M_C_PCH, NACL_5M_1E-3M_C_PCH, NACL_5M_1E-4M_C_PCH, NACL_5M_1E-5M_C_PCH, NACL_5M_PCH $\}$

3. $n \in \{$
- 001 through 009 for ERDA-6_001M_C_PCH
- 001 through 020 for ERDA-6_01M_C_PCH
- 001 through 011 for ERDA-6_1E-4M_C_PCH
- 001 through 012 for ERDA-6_1E-5M_C_PCH
- 001 through 015 for ERDA-6_PCH
- 001 through 016 for GWB_1E-2M_C_PCH
- 001 through 012 for GWB_1E-3M_C_PCH
- 001 through 011 for GWB_1E-4M_C_PCH
- 001 through 014 for GWB_1E-5M_C_PCH
- 001 through 014 for GWB_PCH
- 001 through 033 for NACL_5M_1E-2M_C_PCH
- 001 through 013 for NACL_5M_1E-3M_C_PCH
- 001 through 016 for NACL_5M_1E-4M_C_PCH
- 001 through 016 for NACL_5M_1E-5M_C_PCH
- 001 through 048 for NACL_5M_PCH

$\}$

Table 4. Run-Control Information for the FMT Calculations Conducted for the Comparisons with Rai et al. (1995).

	File Names[1,2]	CMS Library	CMS Class
Script	EVAL_FMT_PABC09.COM	LIBPABC09_EVAL	PABC09-0
Script Input	EVAL_FMT_PABC09_RAI95_*t*_NA2CO3.INP	LIBPABC09_EVAL	PABC09-0
Script Log	EVAL_FMT_PABC09_RAI95_*t*_NA2C03.LOG	LIBPABC09_FMT	PABC09-0
FMT:			
Input	FMT_050405.CHEMDAT	LIBPABC09_FMT	PABC09-0
Input	FMT_GENERIC.RHOMIN	LIBPABC09_FMT	PABC09-0
Input	FMT_PABC09_RAI95_*t*_NA2CO3_*n*.IN	LIBPABC09_FMT	PABC09-0
Input	FMT_PABC09_RAI95_*t*_NA2CO3_*n*.INGUESS	LIBPABC09_FMT	PABC09-0
Output	FMT_PABC09_RAI95_*t*_NA2C03_*n*.OUT	LIBPABC09_FMT	PABC09-0
Output	FMT_PABC09_RAI95_*t*_NA2CO3_*n*.FOR088	Not kept	Not kept

1. t ε {NAOH_1M, 1E-1M_NAOH}
2. n ε { 001 through 015 for NAOH_1M and 001 through 005 for 1E-1M_NAOH}

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3 RESULTS

This section presents the results of our analyses for three closely related EPA comments arising from its completeness review of the CRA-2009 (U.S. DOE, 2009) for the WIPP.

3.1 Comment 8 of the Follow-Up Comments to 1-23-6

Comment 8 of the EPA's January 25, 2010, follow-up comments to the issues first raised in Comment 1-23-6 of its May 21, 2009 letter to the DOE (Cotsworth, 2009) requested that we "perform FMT calculations to predict the concentrations of Nd(III) under the conditions of the Nd(III) solubility experiments in the NaCl, GWB, and ERDA-6 brines."

Tables 5 through 19 provide the final measured values of the pcH, the initial CO_3^{2-} concentrations, the run type, the final, measured Nd(III) concentrations (solubilities) provided by Borkowski (2010, Tables 2–8), the Nd(III) solubilities predicted by FMT (this analysis), and the FMT run numbers. For each solution, we combined all of the runs with different initial CO_3^{2-} concentrations (0, 1×10^{-5} M, 1×10^{-4} M, 1×10^{-3} M, and 1×10^{-5} M) in one table in order of increasing pcH to facilitate locating each experiment to add the solubilities predicted by FMT and checking this and the other information in these tables. If two runs had the same final pcH, we arranged them in order of increasing initial CO_3^{2-} concentration. If two runs had the same final pcH and initial CO_3^{2-} concentration, we arranged them in order of increasing measured Nd(III) solubility.

Figures 1 through 15 compare the measured and predicted Nd(III) solubilities as a function of the final measured values of the pcH. Each figure includes all of the measured and predicted Nd(III) solubilities obtained for a given initial CO_3^{2-} concentrations. Therefore, there are five figures for each solution (5 M NaCl, GWB, and ERDA-6).

3.1.1 Results for Borkowski et al. (2009), 5 M NaCl

Table 5 and Figure 1 demonstrate that, for the CO_3^{2-}-free 5 M NaCl solutions, FMT overpredicted most of the Nd(III) solubilities measured at final pcH values ≤ 10.66, with the exceptions of four solubilities measured at pcH = 8.25, 8.29, 9.85, and 10.63. However, FMT underpredicted the solubilities measured at pcH > 10.66.

Table 6 and Figure 2 show that, for the 5 M NaCl solutions with an initial CO_3^{2-} concentration of 1×10^{-5} M, FMT overpredicted the Nd(III) solubilities measured over the entire range of final pcH values, with the exception of one at pcH = 9.22.

According to Table 7 and Figure 3, FMT overpredicted the Nd(III) solubilities measured over the entire range of final pcH values for 5 M NaCl solutions with an initial CO_3^{2-} concentration of 1×10^{-4} M.

Table 5. Comparison of the 36 Nd(III) Solubilities Measured in CO_3^{2-}-Free 5 M NaCl (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 1 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.19	Undersaturation	1.02×10^{-6}	2.59×10^{-6}	FMT_LANL09_NACL_5M_PCH_001
8.21	Undersaturation	4.51×10^{-8}	2.45×10^{-6}	FMT_LANL09_NACL_5M_PCH_002
8.25	Undersaturation	5.07×10^{-6}	2.20×10^{-6}	FMT_LANL09_NACL_5M_PCH_003
8.29	Undersaturation	3.13×10^{-6}	1.95×10^{-6}	FMT_LANL09_NACL_5M_PCH_005
8.29	Undersaturation	3.49×10^{-9}	1.95×10^{-6}	FMT_LANL09_NACL_5M_PCH_005
8.70	Oversaturation	1.67×10^{-7}	6.89×10^{-7}	FMT_LANL09_NACL_5M_PCH_007
8.80	Oversaturation	4.47×10^{-8}	5.40×10^{-7}	FMT_LANL09_NACL_5M_PCH_008
9.06	Oversaturation	1.12×10^{-7}	2.92×10^{-7}	FMT_LANL09_NACL_5M_PCH_011
9.09	Undersaturation	1.99×10^{-8}	2.71×10^{-7}	FMT_LANL09_NACL_5M_PCH_012
9.11	Oversaturation	3.60×10^{-8}	2.60×10^{-7}	FMT_LANL09_NACL_5M_PCH_013
9.15	Undersaturation	6.92×10^{-9}	2.36×10^{-7}	FMT_LANL09_NACL_5M_PCH_014
9.57	Oversaturation	1.11×10^{-8}	8.85×10^{-8}	FMT_LANL09_NACL_5M_PCH_016
9.58	Undersaturation	5.78×10^{-9}	8.65×10^{-8}	FMT_LANL09_NACL_5M_PCH_017
9.62	Undersaturation	2.99×10^{-8}	7.93×10^{-8}	FMT_LANL09_NACL_5M_PCH_018.
9.64	Undersaturation	8.63×10^{-9}	7.55×10^{-8}	FMT_LANL09_NACL_5M_PCH_019
9.67	Oversaturation	9.36×10^{-9}	7.06×10^{-8}	FMT_LANL09_NACL_5M_PCH_020
9.69	Undersaturation	5.91×10^{-8}	6.75×10^{-8}	FMT_LANL09_NACL_5M_PCH_021
9.85	Undersaturation	4.93×10^{-8}	4.70×10^{-8}	FMT_LANL09_NACL_5M_PCH_024
10.06	Undersaturation	1.26×10^{-8}	2.91×10^{-8}	FMT_LANL09_NACL_5M_PCH_025
10.63	Oversaturation	1.50×10^{-8}	8.13×10^{-9}	FMT_LANL09_NACL_5M_PCH_029
10.66	Oversaturation	6.60×10^{-9}	7.70×10^{-9}	FMT_LANL09_NACL_5M_PCH_030

Table 5 continued on next page

Table 5. Comparison of the 36 Nd(III) Solubilities Measured in CO_3^{2-}-Free 5 M NaCl (Borkowski et al., 2009) with Those Predicted by FMT (this analysis) (continued).

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
11.51	Undersaturation	1.73×10^{-8}	1.50×10^{-9}	FMT_LANL09_NACL_5M_PCH_033
11.54	Undersaturation	1.28×10^{-8}	1.43×10^{-9}	FMT_LANL09_NACL_5M_PCH_034
11.58	Oversaturation	7.21×10^{-9}	1.35×10^{-9}	FMT_LANL09_NACL_5M_PCH_037
11.61	Oversaturation	9.43×10^{-9}	1.29×10^{-9}	FMT_LANL09_NACL_5M_PCH_038
11.77	Undersaturation	5.37×10^{-9}	1.05×10^{-9}	FMT_LANL09_NACL_5M_PCH_039
11.77	Undersaturation	7.99×10^{-9}	1.05×10^{-9}	FMT_LANL09_NACL_5M_PCH_039
11.82	Oversaturation	4.93×10^{-9}	9.87×10^{-10}	FMT_LANL09_NACL_5M_PCH_040
11.84	Oversaturation	7.31×10^{-9}	9.65×10^{-10}	FMT_LANL09_NACL_5M_PCH_041
12.70	Undersaturation	7.69×10^{-9}	5.57×10^{-10}	FMT_LANL09_NACL_5M_PCH_044
12.71	Undersaturation	6.14×10^{-9}	5.55×10^{-10}	FMT_LANL09_NACL_5M_PCH_045
12.71	Oversaturation	6.78×10^{-9}	5.55×10^{-10}	FMT_LANL09_NACL_5M_PCH_045
12.71	Oversaturation	9.38×10^{-9}	5.55×10^{-10}	FMT_LANL09_NACL_5M_PCH_045
12.98	Oversaturation	7.24×10^{-9}	5.27×10^{-10}	FMT_LANL09_NACL_5M_PCH_046
12.99	Oversaturation	9.48×10^{-9}	5.26×10^{-10}	FMT_LANL09_NACL_5M_PCH_047
13.00	Undersaturation	3.06×10^{-9}	5.25×10^{-10}	FMT_LANL09_NACL_5M_PCH_048

Table 6. Comparison of the 16 Nd(III) Solubilities Measured in 5 M NaCl with an Initial CO_3^{2} Concentration of 1×10^{-5} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 2 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.14	Oversaturation	1.60×10^{-7}	1.75×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_016
8.26	Oversaturation	2.32×10^{-7}	1.65×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_001
8.29	Undersaturation	8.77×10^{-8}	1.64×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_002
8.31	Oversaturation	3.67×10^{-7}	1.63×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_003
8.35	Oversaturation	3.38×10^{-7}	1.61×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_004
8.42	Undersaturation	1.05×10^{-9}	1.40×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_005
8.46	Undersaturation	1.99×10^{-9}	1.27×10^{-6}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_006
8.57	Oversaturation	8.22×10^{-9}	9.59×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_007
8.58	Oversaturation	1.19×10^{-9}	9.35×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_008
8.78	Undersaturation	4.13×10^{-9}	5.73×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_009
8.87	Undersaturation	1.02×10^{-8}	4.62×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_010
8.92	Oversaturation	6.48×10^{-9}	4.10×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_011
8.94	Oversaturation	7.44×10^{-9}	3.91×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_012
9.22	Undersaturation	2.45×10^{-7}	2.01×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_013
9.24	Undersaturation	1.61×10^{-8}	1.92×10^{-7}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_014
10.80	Undersaturation	4.22×10^{-9}	5.69×10^{-9}	FMT_LANL09_NACL_5M_1E-5M_C_PCH_015

Table 7. Comparisons of the 16 Nd(III) Solubilities Measured in 5 M NaCl with an Initial CO_3^{2-} Concentration of 1×10^{-4} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 3 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.34	Undersaturation	1.20×10^{-8}	1.95×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_001
8.35	Oversaturation	1.18×10^{-7}	1.94×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_002
8.42	Undersaturation	8.68×10^{-9}	1.90×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_003
8.47	Oversaturation	6.40×10^{-8}	1.87×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_004
8.51	Oversaturation	6.00×10^{-9}	1.85×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_005
8.57	Oversaturation	4.09×10^{-9}	1.83×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_006
8.61	Oversaturation	5.62×10^{-9}	1.83×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_007
8.63	Undersaturation	6.92×10^{-9}	1.82×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_008
8.64	Undersaturation	7.85×10^{-9}	1.82×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_009
8.75	Oversaturation	3.11×10^{-9}	1.81×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_010
9.10	Oversaturation	9.86×10^{-9}	1.91×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_011
9.11	Undersaturation	8.24×10^{-9}	1.92×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_012
9.12	Undersaturation	1.22×10^{-8}	1.92×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_013
9.14	Oversaturation	1.53×10^{-8}	1.93×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_014
9.27	Undersaturation	1.73×10^{-8}	1.81×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_015
9.39	Undersaturation	2.39×10^{-8}	1.37×10^{-7}	FMT_LANL09_NACL_5M_1E-4M_C_PCH_016

Table 8. Comparisons of the 16 Nd(III) Solubilities Measured in 5 M NaCl with an Initial CO_3^{2-} Concentration of 1×10^{-3} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 4 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.69	Undersaturation	4.20×10^{-9}	2.94×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_001
8.71	Oversaturation	6.74×10^{-9}	2.93×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_002
8.82	Oversaturation	3.58×10^{-9}	2.88×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_003
8.84	Oversaturation	4.86×10^{-9}	2.87×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_004
8.89	Oversaturation	3.42×10^{-9}	2.86×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_005
8.89	Oversaturation	7.03×10^{-9}	2.86×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_005
8.93	Undersaturation	1.66×10^{-8}	2.86×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_006
9.00	Undersaturation	8.90×10^{-9}	2.87×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_007
9.00	Undersaturation	1.31×10^{-8}	2.87×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_007
9.09	Undersaturation	6.39×10^{-9}	2.90×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_008
9.09	Oversaturation	6.78×10^{-9}	2.90×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_008
9.10	Oversaturation	1.38×10^{-8}	2.90×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_009
9.13	Undersaturation	5.05×10^{-9}	2.92×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_010
9.15	Oversaturation	8.13×10^{-9}	2.93×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_011
9.16	Undersaturation	2.79×10^{-9}	2.94×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_012
9.26	Undersaturation	2.37×10^{-8}	3.01×10^{-8}	FMT_LANL09_NACL_5M_1E-3M_C_PCH_013

Table 9. Comparisons of the 36 Nd(III) Solubilities Measured in 5 M NaCl with an Initial CO_3^{2-} Concentration of 1×10^{-2} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 5 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.54	Oversaturation	2.79×10^{-8}	1.13×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_001
8.71	Oversaturation	2.25×10^{-8}	1.37×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_002
8.85	Undersaturation	2.22×10^{-8}	1.68×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_003
8.85	Undersaturation	2.74×10^{-8}	1.68×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_003
9.07	Undersaturation	6.95×10^{-8}	2.45×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_004
9.22	Undersaturation	5.12×10^{-8}	3.28×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_005
9.23	Undersaturation	6.48×10^{-8}	3.35×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_006
9.24	Undersaturation	7.01×10^{-8}	3.42×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_007
9.25	Oversaturation	8.04×10^{-8}	3.49×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_008
9.32	Oversaturation	3.74×10^{-8}	4.06×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_009
9.36	Oversaturation	4.29×10^{-8}	4.43×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_010
9.36	Oversaturation	4.40×10^{-8}	4.43×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_010
9.48	Oversaturation	8.69×10^{-8}	5.36×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_011
9.50	Undersaturation	1.05×10^{-7}	5.45×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_012
9.51	Undersaturation	2.06×10^{-7}	5.50×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_013
9.54	Undersaturation	1.89×10^{-7}	5.63×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_014
9.56	Undersaturation	2.19×10^{-7}	5.72×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_015
9.59	Oversaturation	8.14×10^{-8}	5.84×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_016
9.63	Oversaturation	8.95×10^{-8}	5.99×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_017
9.66	Oversaturation	7.47×10^{-8}	6.09×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_018

Table 9 continued on next page

Table 9. Comparisons of the 36 Nd(III) Solubilities Measured in 5 M NaCl with an Initial CO_3^{2-} Concentration of 1×10^{-2} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis) (continued). See Figure 5 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
9.66	Oversaturation	1.30×10^{-7}	6.09×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_018
9.69	Undersaturation	1.03×10^{-7}	6.18×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_019
9.70	Oversaturation	9.98×10^{-8}	6.21×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_020
9.83	Oversaturation	1.42×10^{-7}	6.44×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_021
9.84	Undersaturation	1.39×10^{-7}	6.45×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_022
9.88	Oversaturation	9.40×10^{-8}	6.46×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_023
9.90	Undersaturation	1.10×10^{-7}	6.45×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_024
10.05	Undersaturation	1.24×10^{-7}	6.22×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_025
10.06	Undersaturation	1.27×10^{-7}	6.19×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_026
10.15	Oversaturation	1.63×10^{-7}	5.88×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_027
10.30	Oversaturation	2.01×10^{-7}	5.18×10^{-7}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_028
11.91	Undersaturation	2.85×10^{-8}	9.51×10^{-10}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_029
13.05	Undersaturation	1.83×10^{-8}	5.29×10^{-10}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_030
13.14	Oversaturation	1.90×10^{-8}	5.23×10^{-10}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_031
13.16	Oversaturation	2.29×10^{-8}	5.22×10^{-10}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_032
13.17	Undersaturation	2.22×10^{-8}	5.22×10^{-10}	FMT_LANL09_NACL_5M_1E-2M_C_PCH_033

Table 10. Comparison of the 20 Nd(III) Solubilities Measured in CO_3^{2-}-Free ERDA-6 (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 6 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
7.55	Undersaturation	1.43×10^{-6}	2.28×10^{-5}	FMT_LANL09_ERDA-6_PCH_001
7.55	Undersaturation	1.45×10^{-6}	2.28×10^{-5}	FMT_LANL09_ERDA-6_PCH_001
7.66	Oversaturation	1.74×10^{-7}	1.25×10^{-5}	FMT_LANL09_ERDA-6_PCH_002
7.67	Oversaturation	1.47×10^{-7}	1.19×10^{-5}	FMT_LANL09_ERDA-6_PCH_003
8.46	Oversaturation	2.18×10^{-8}	5.67×10^{-7}	FMT_LANL09_ERDA-6_PCH_004
8.46	Undersaturation	4.87×10^{-8}	5.67×10^{-7}	FMT_LANL09_ERDA-6_PCH_004
8.46	Undersaturation	6.38×10^{-8}	5.67×10^{-7}	FMT_LANL09_ERDA-6_PCH_004
8.47	Oversaturation	3.19×10^{-8}	5.52×10^{-7}	FMT_LANL09_ERDA-6_PCH_005
9.26	Undersaturation	6.15×10^{-8}	7.64×10^{-8}	FMT_LANL09_ERDA-6_PCH_006
9.26	Oversaturation	9.65×10^{-8}	7.64×10^{-8}	FMT_LANL09_ERDA-6_PCH_006
9.26	Undersaturation	4.19×10^{-7}	7.64×10^{-8}	FMT_LANL09_ERDA-6_PCH_006
9.27	Oversaturation	1.00×10^{-7}	7.46×10^{-8}	FMT_LANL09_ERDA-6_PCH_007
10.29	Undersaturation	1.94×10^{-7}	7.30×10^{-9}	FMT_LANL09_ERDA-6_PCH_008
10.31	Undersaturation	3.52×10^{-8}	6.97×10^{-9}	FMT_LANL09_ERDA-6_PCH_009
10.32	Oversaturation	6.88×10^{-8}	6.82×10^{-9}	FMT_LANL09_ERDA-6_PCH_010
10.33	Oversaturation	1.31×10^{-7}	6.71×10^{-9}	FMT_LANL09_ERDA-6_PCH_015
10.55	Oversaturation	1.03×10^{-8}	4.16×10^{-9}	FMT_LANL09_ERDA-6_PCH_011
10.56	Oversaturation	8.04×10^{-9}	4.07×10^{-9}	FMT_LANL09_ERDA-6_PCH_012
10.58	Undersaturation	2.07×10^{-8}	3.91×10^{-9}	FMT_LANL09_ERDA-6_PCH_013
10.62	Undersaturation	1.80×10^{-8}	3.59×10^{-9}	FMT_LANL09_ERDA-6_PCH_014

Table 11. Comparison of the 12 Nd(III) Solubilities Measured in ERDA-6 with an Initial CO_3^{2-} Concentration of 1×10^{-5} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 7 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.05	Oversaturation	4.54×10^{-8}	1.39×10^{-6}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_002
8.08	Undersaturation	4.04×10^{-8}	1.34×10^{-6}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_003
8.12	Undersaturation	1.86×10^{-8}	1.28×10^{-6}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_004
8.96	Oversaturation	5.05×10^{-8}	1.59×10^{-7}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_005
8.98	Undersaturation	1.20×10^{-7}	1.51×10^{-7}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_006
8.99	Oversaturation	3.56×10^{-8}	1.48×10^{-7}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_007
9.00	Oversaturation	4.86×10^{-8}	1.44×10^{-7}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_008
9.01	Undersaturation	5.11×10^{-8}	1.41×10^{-7}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_009
9.47	Undersaturation	7.19×10^{-7}	4.89×10^{-8}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_010
9.61	Undersaturation	8.30×10^{-7}	3.68×10^{-8}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_011
9.73	Undersaturation	8.65×10^{-7}	3.03×10^{-8}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_012
10.05	Undersaturation	7.12×10^{-7}	3.74×10^{-8}	FMT_LANL09_ERDA-6_1E-5M_C_PCH_001

Table 12. Comparison of the 12 Nd(III) Solubilities Measured in ERDA-6 with an Initial CO_3^{2-} Concentration of 1×10^{-4} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 8 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
7.97	Undersaturation	4.14×10^{-8}	1.95×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_002
7.97	Undersaturation	7.51×10^{-8}	1.95×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_002
7.98	Oversaturation	1.79×10^{-7}	1.92×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_003
7.99	Oversaturation	1.78×10^{-7}	1.88×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_004
8.98	Oversaturation	2.58×10^{-8}	1.20×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_005
9.00	Oversaturation	4.25×10^{-8}	1.21×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_006
9.01	Undersaturation	5.24×10^{-8}	1.21×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_007
9.03	Undersaturation	5.86×10^{-8}	1.22×10^{-7}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_008
9.66	Undersaturation	5.59×10^{-7}	3.37×10^{-8}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_009
9.70	Undersaturation	6.32×10^{-7}	3.16×10^{-8}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_010
9.80	Undersaturation	6.34×10^{-7}	2.82×10^{-8}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_011
10.13	Undersaturation	6.25×10^{-7}	5.18×10^{-8}	FMT_LANL09_ERDA-6_1E-4M_C_PCH_001

Table 13. Comparison of the 12 Nd(III) Solubilities Measured in ERDA-6 with an Initial CO_3^{2-} Concentration of 1×10^{-3} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 9 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.06	Undersaturation	3.06×10^{-8}	3.68×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_001
8.06	Oversaturation	8.41×10^{-8}	3.68×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_001
8.08	Undersaturation	3.63×10^{-8}	3.57×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_002
8.08	Oversaturation	7.89×10^{-8}	3.57×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_002
9.03	Oversaturation	6.03×10^{-8}	2.05×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_003
9.04	Undersaturation	4.73×10^{-8}	2.05×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_004
9.04	Oversaturation	5.63×10^{-8}	2.05×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_004
9.06	Undersaturation	4.08×10^{-8}	2.05×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_005
9.44	Undersaturation	1.01×10^{-7}	2.32×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_006
9.61	Undersaturation	6.19×10^{-7}	2.60×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_007
9.85	Undersaturation	5.48×10^{-7}	2.75×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_008
9.90	Undersaturation	5.65×10^{-7}	2.78×10^{-8}	FMT_LANL09_ERDA-6_001M_C_PCH_009

Table 14. Comparison of the 22 Nd(III) Solubilities Measured in ERDA-6 with an Initial CO_3^{2-} Concentration of 1×10^{-2} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 10 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
8.2	Oversaturation	2.00×10^{-8}	9.04×10^{-8}	FMT_LANL09_ERDA-6_01M_C_PCH_003
8.21	Oversaturation	2.32×10^{-8}	9.08×10^{-8}	FMT_LANL09_ERDA-6_01M_C_PCH_004
8.23	Undersaturation	2.21×10^{-8}	9.17×10^{-8}	FMT_LANL09_ERDA-6_01M_C_PCH_005
8.25	Undersaturation	2.22×10^{-8}	9.26×10^{-8}	FMT_LANL09_ERDA-6_01M_C_PCH_006
8.56	Oversaturation	2.13×10^{-8}	1.21×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_007
8.57	Undersaturation	2.45×10^{-8}	1.22×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_008
8.60	Undersaturation	1.15×10^{-8}	1.27×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_009
8.61	Oversaturation	1.93×10^{-8}	1.29×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_010
8.98	Oversaturation	2.89×10^{-8}	2.30×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_011
8.99	Oversaturation	2.58×10^{-8}	2.34×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_012
9.00	Undersaturation	3.19×10^{-8}	2.38×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_013
9.31	Oversaturation	7.97×10^{-9}	3.10×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_014
9.48	Undersaturation	1.88×10^{-8}	3.24×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_015
9.50	Undersaturation	3.31×10^{-8}	3.25×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_016
9.61	Oversaturation	5.03×10^{-8}	3.22×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_017
9.75	Undersaturation	5.28×10^{-8}	3.06×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_018
9.87	Oversaturation	3.18×10^{-8}	2.83×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_019
9.90	Undersaturation	2.96×10^{-8}	2.76×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_020
10.0	Oversaturation	5.51×10^{-8}	2.52×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_001
10.3	Oversaturation	3.93×10^{-8}	2.30×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_002
10.3	Undersaturation	6.40×10^{-8}	2.30×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_002
10.3	Undersaturation	1.19×10^{-7}	2.30×10^{-7}	FMT_LANL09_ERDA-6_01M_C_PCH_002

Table 15. Comparison of the 16 Nd(III) Solubilities Measured in CO_3^{2-}-Free GWB (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 11 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
6.6	Undersaturation	5.27×10^{-6}	2.17×10^{-2}	FMT_LANL09_GWB_PCH_001
6.64	Undersaturation	3.59×10^{-6}	1.67×10^{-2}	FMT_LANL09_GWB_PCH_002
6.67	Oversaturation	4.48×10^{-6}	1.39×10^{-2}	FMT_LANL09_GWB_PCH_003
6.68	Oversaturation	4.76×10^{-6}	1.30×10^{-2}	FMT_LANL09_GWB_PCH_004
7.29	Oversaturation	1.27×10^{-7}	2.37×10^{-4}	FMT_LANL09_GWB_PCH_005
7.30	Undersaturation	8.98×10^{-7}	2.22×10^{-4}	FMT_LANL09_GWB_PCH_006
7.30	Undersaturation	9.81×10^{-7}	2.22×10^{-4}	FMT_LANL09_GWB_PCH_006
7.32	Oversaturation	1.43×10^{-7}	1.97×10^{-4}	FMT_LANL09_GWB_PCH_007
7.95	Oversaturation	7.36×10^{-8}	7.70×10^{-6}	FMT_LANL09_GWB_PCH_008
7.96	Undersaturation	4.56×10^{-7}	7.44×10^{-6}	FMT_LANL09_GWB_PCH_009
7.96	Undersaturation	5.21×10^{-7}	7.44×10^{-6}	FMT_LANL09_GWB_PCH_009
7.97	Oversaturation	5.34×10^{-8}	7.15×10^{-6}	FMT_LANL09_GWB_PCH_010
8.58	Undersaturation	8.05×10^{-7}	1.15×10^{-6}	FMT_LANL09_GWB_PCH_011
8.59	Undersaturation	5.75×10^{-7}	1.12×10^{-6}	FMT_LANL09_GWB_PCH_012
8.62	Oversaturation	3.93×10^{-7}	1.04×10^{-6}	FMT_LANL09_GWB_PCH_013
8.63	Oversaturation	4.14×10^{-7}	1.02×10^{-6}	FMT_LANL09_GWB_PCH_014

Table 16. Comparison of the 14 Nd(III) Solubilities Measured in GWB with an Initial CO_3^{2-} Concentration of 1×10^{-5} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 12 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
6.64	Undersaturation	2.72×10^{-6}	6.18×10^{-5}	FMT_LANL09_GWB_1E-5M_C_PCH_001
6.68	Undersaturation	2.33×10^{-6}	5.55×10^{-5}	FMT_LANL09_GWB_1E-5M_C_PCH_002
7.28	Undersaturation	5.82×10^{-8}	1.13×10^{-5}	FMT_LANL09_GWB_1E-5M_C_PCH_003
7.35	Undersaturation	5.45×10^{-8}	9.49×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_004
7.40	Oversaturation	1.40×10^{-7}	8.39×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_005
7.43	Oversaturation	1.31×10^{-7}	7.82×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_006
7.80	Oversaturation	2.03×10^{-7}	3.84×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_007
7.83	Oversaturation	2.08×10^{-7}	3.70×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_008
7.88	Undersaturation	8.64×10^{-8}	3.51×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_009
7.89	Undersaturation	1.18×10^{-7}	3.48×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_010
8.30	Oversaturation	7.15×10^{-7}	2.44×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_011
8.32	Oversaturation	8.14×10^{-7}	2.30×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_012
8.38	Undersaturation	3.48×10^{-7}	1.95×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_013
8.39	Undersaturation	3.12×10^{-7}	1.89×10^{-6}	FMT_LANL09_GWB_1E-5M_C_PCH_014

Table 17. Comparison of the 14 Nd(III) Solubilities Measured in GWB with an Initial CO_3^{2-} Concentration of 1×10^{-4} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 13 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
6.63	Undersaturation	2.18×10^{-6}	3.49×10^{-5}	FMT_LANL09_GWB_1E-4M_C_PCH_001
6.63	Undersaturation	4.56×10^{-6}	3.49×10^{-5}	FMT_LANL09_GWB_1E-4M_C_PCH_001
7.15	Undersaturation	1.23×10^{-7}	4.05×10^{-6}	FMT_LANL09_GWB_1E-4M_C_PCH_002
7.17	Undersaturation	1.28×10^{-7}	3.74×10^{-6}	FMT_LANL09_GWB_1E-4M_C_PCH_003
7.32	Oversaturation	2.06×10^{-7}	2.10×10^{-6}	FMT_LANL09_GWB_1E-4M_C_PCH_004
7.33	Oversaturation	2.08×10^{-7}	2.03×10^{-6}	FMT_LANL09_GWB_1E-4M_C_PCH_005
7.80	Undersaturation	8.14×10^{-8}	5.57×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_006
7.87	Oversaturation	1.85×10^{-7}	5.02×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_007
7.87	Oversaturation	1.88×10^{-7}	5.02×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_007
7.88	Undersaturation	8.24×10^{-8}	4.96×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_008
8.36	Undersaturation	4.12×10^{-7}	4.58×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_009
8.36	Oversaturation	8.85×10^{-7}	4.58×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_009
8.37	Oversaturation	8.38×10^{-7}	4.61×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_010
8.38	Undersaturation	3.38×10^{-7}	4.65×10^{-7}	FMT_LANL09_GWB_1E-4M_C_PCH_011

Table 18. Comparison of the 14 Nd(III) Solubilities Measured in GWB with an Initial CO_3^{2-} Concentration of 1×10^{-3} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 14 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
6.58	Undersaturation	1.83×10^{-6}	6.47×10^{-6}	FMT_LANL09_GWB_1E-3M_C_PCH_001
6.62	Undersaturation	1.77×10^{-6}	5.34×10^{-6}	FMT_LANL09_GWB_1E-3M_C_PCH_002
7.32	Undersaturation	6.73×10^{-8}	3.15×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_003
7.33	Undersaturation	5.83×10^{-8}	3.05×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_004
7.40	Oversaturation	1.01×10^{-7}	2.50×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_005
7.42	Oversaturation	9.49×10^{-8}	2.36×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_006
7.82	Undersaturation	1.20×10^{-7}	1.34×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_007
7.85	Undersaturation	8.87×10^{-8}	1.34×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_008
7.87	Oversaturation	1.63×10^{-7}	1.34×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_009
7.88	Oversaturation	1.76×10^{-7}	1.34×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_010
8.34	Undersaturation	5.85×10^{-7}	1.83×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_011
8.34	Oversaturation	3.12×10^{-6}	1.83×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_011
8.39	Undersaturation	5.80×10^{-7}	1.89×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_012
8.39	Oversaturation	2.34×10^{-6}	1.89×10^{-7}	FMT_LANL09_GWB_1E-3M_C_PCH_012

Table 19. Comparison of the 16 Nd(III) Solubilities Measured in GWB with an Initial CO_3^{2-} Concentration of 1×10^{-2} M (Borkowski et al., 2009) with Those Predicted by FMT (this analysis). See Figure 15 (this report) for the scatter plot of measured and predicted solubilities.

Final pC_H	Type of Experiment	Nd(III) Solubility, Measured (M)	Nd(III) Solubility, Predicted (M)	FMT Run Number
6.54	Oversaturation	8.87×10^{-6}	1.47×10^{-6}	FMT_LANL09_GWB_1E-2M_C_PCH_001
6.74	Oversaturation	1.09×10^{-5}	1.06×10^{-6}	FMT_LANL09_GWB_1E-2M_C_PCH_002
6.78	Undersaturation	2.08×10^{-6}	1.08×10^{-6}	FMT_LANL09_GWB_1E-2M_C_PCH_003
6.79	Undersaturation	8.41×10^{-7}	1.09×10^{-6}	FMT_LANL09_GWB_1E-2M_C_PCH_004
7.46	Oversaturation	2.83×10^{-7}	1.25×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_005
7.51	Oversaturation	2.15×10^{-7}	1.51×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_006
7.59	Undersaturation	7.82×10^{-8}	2.02×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_007
7.59	Undersaturation	8.44×10^{-8}	2.02×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_007
7.88	Oversaturation	3.77×10^{-7}	4.87×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_008
8.03	Oversaturation	3.07×10^{-7}	6.82×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_009
8.05	Undersaturation	7.38×10^{-8}	7.07×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_010
8.08	Undersaturation	5.14×10^{-8}	7.49×10^{-5}	FMT_LANL09_GWB_1E-2M_C_PCH_011
8.57	Oversaturation	3.00×10^{-6}	1.14×10^{-4}	FMT_LANL09_GWB_1E-2M_C_PCH_013 FMT_LANL09_GWB_1E-2M_C_PCH_015
8.58	Oversaturation	2.68×10^{-6}	1.14×10^{-4}	FMT_LANL09_GWB_1E-2M_C_PCH_012
8.64	Undersaturation	1.74×10^{-6}	1.13×10^{-4}	FMT_LANL09_GWB_1E-2M_C_PCH_014 FMT_LANL09_GWB_1E-2M_C_PCH_016
8.64	Undersaturation	2.34×10^{-6}	1.13×10^{-4}	FMT_LANL09_GWB_1E-2M_C_PCH_014, FMT_LANL09_GWB_1E-2M_C_PCH_016

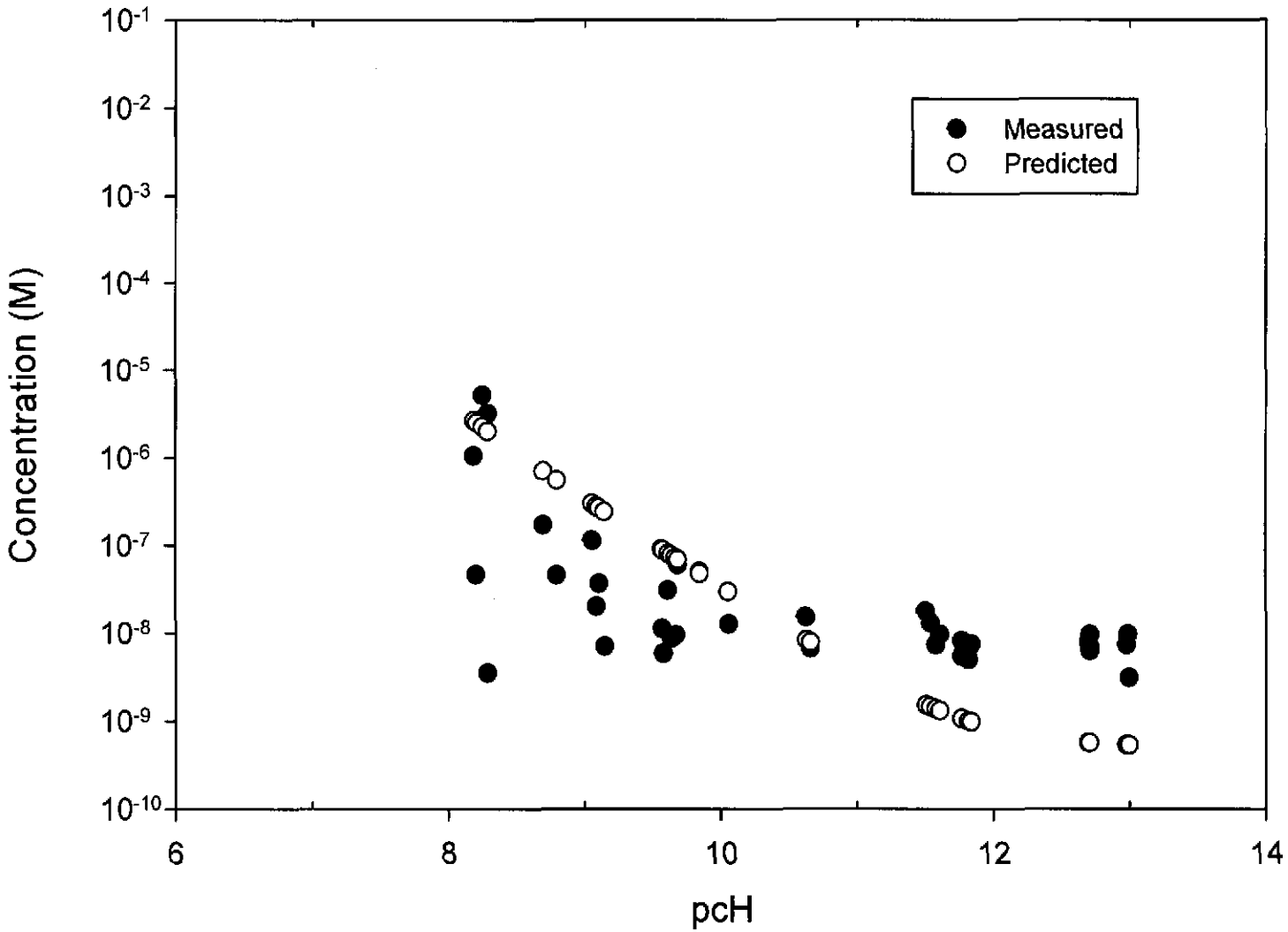

Figure 1. Comparison of the 36 Nd(III) solubilities measured in CO_3^{2-}-free 5 M NaCl (Borkowski et al., 2009) with those predicted by FMT (Table 5, this report).

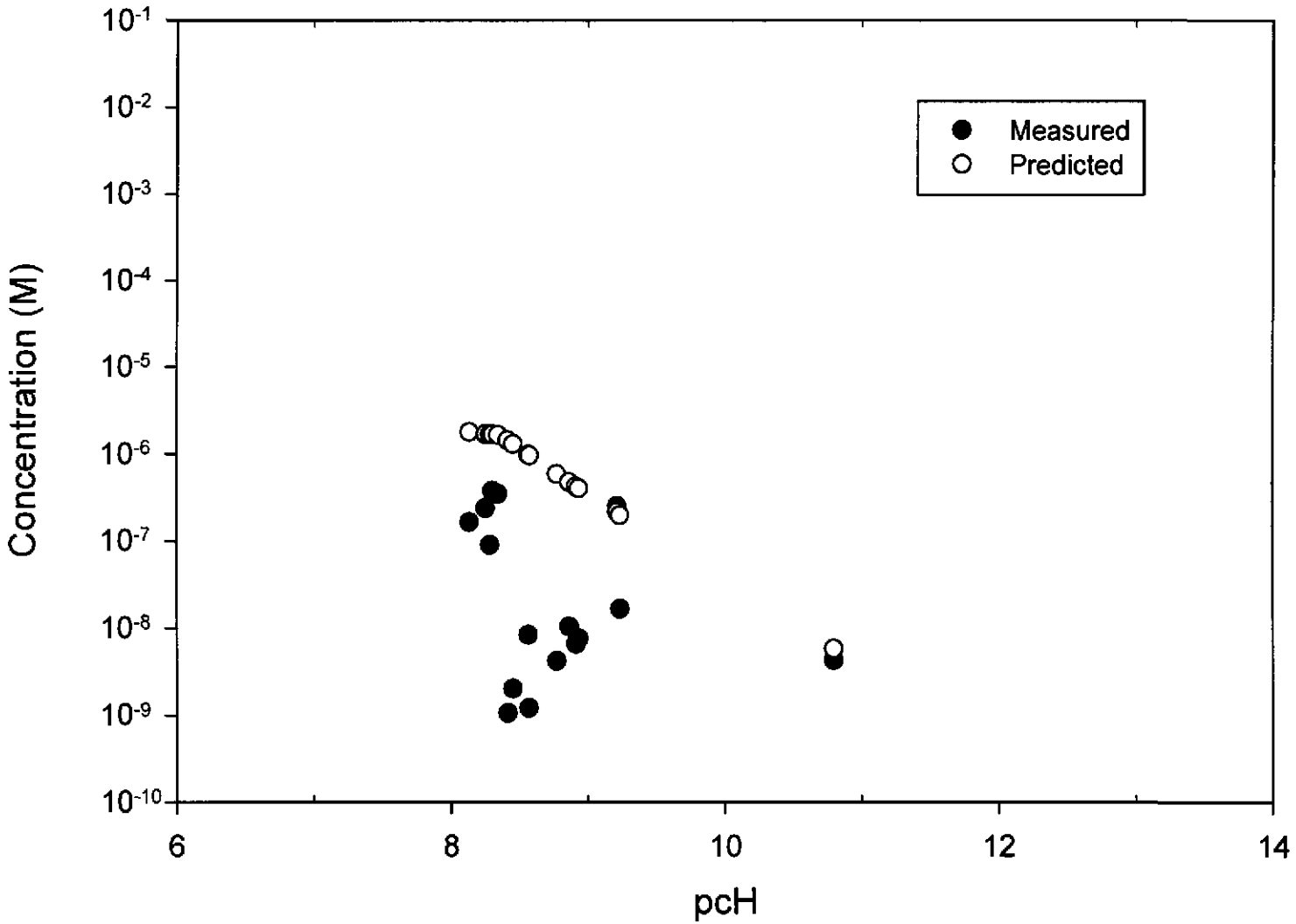

Figure 2. Comparison of the 16 Nd(III) solubilities measured in 5 M NaCl with an initial CO_3^{2-} concentration of 1×10^{-5} M (Borkowski et al., 2009) with those predicted by FMT (Table 6, this report).

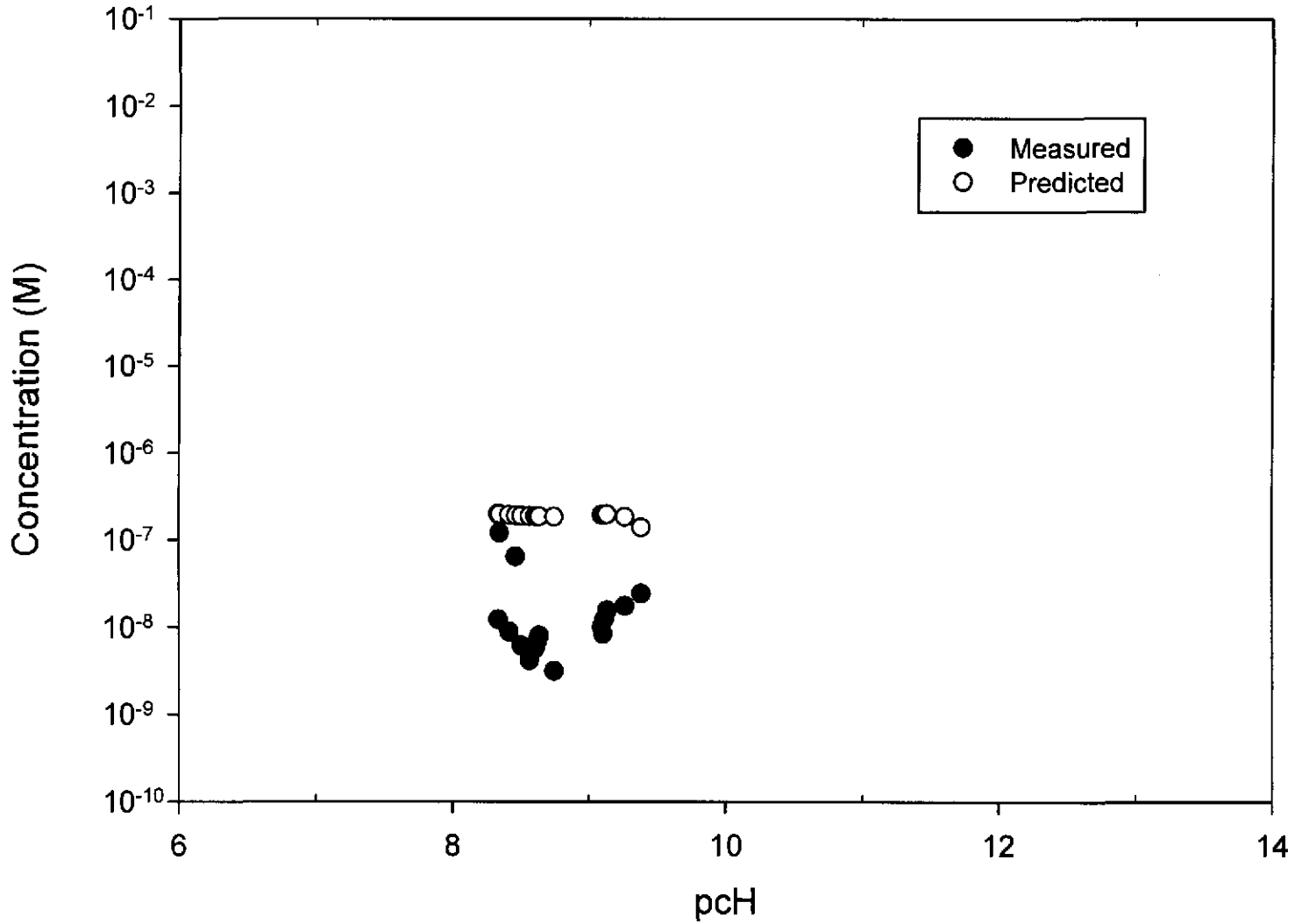

Figure 3. Comparison of the 16 Nd(III) solubilities measured in 5 M NaCl with an initial CO_3^{2-} concentration of 1×10^{-4} M (Borkowski et al., 2009) with those predicted by FMT (Table 7, this report).

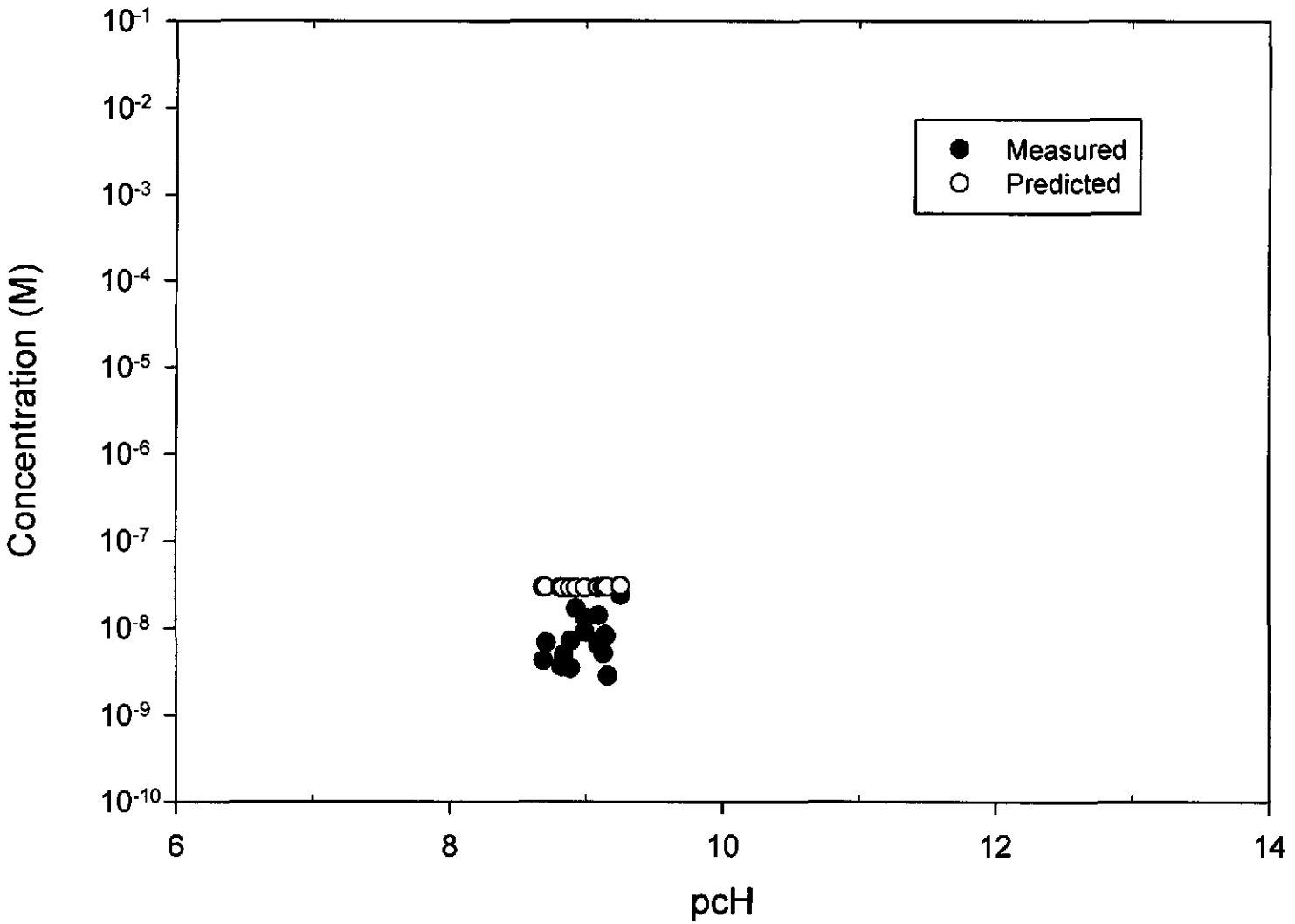

Figure 4. Comparison of the 16 Nd(III) solubilities measured in 5 M NaCl with an initial CO_3^{2-} concentration of 1×10^{-3} M (Borkowski et al., 2009) with those predicted by FMT (Table 8, this report).

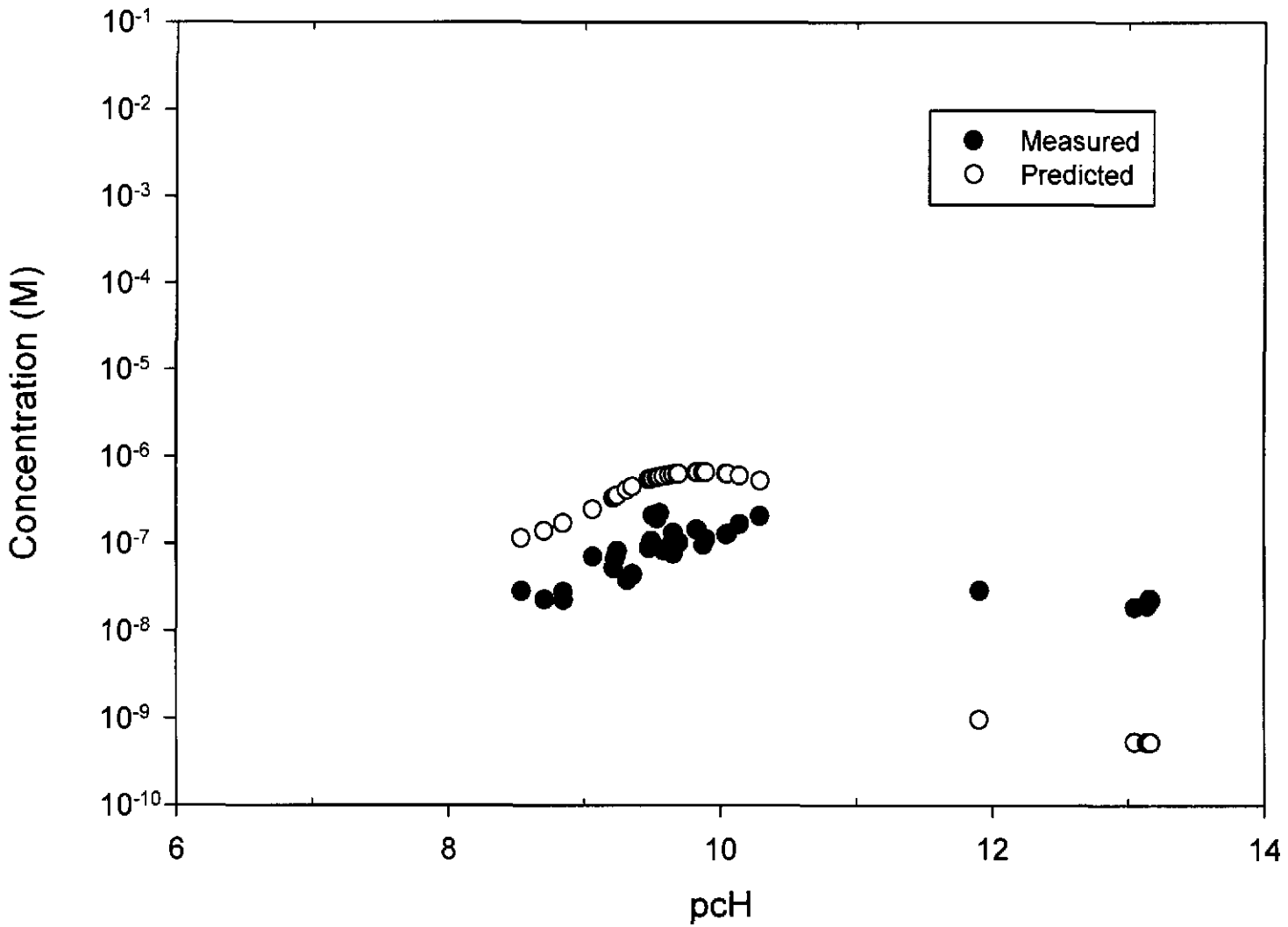

Figure 5. Comparison of the 36 Nd(III) solubilities measured in 5 M NaCl with an initial CO_3^{2-} concentration of 1×10^{-2} M (Borkowski et al., 2009) with those predicted by FMT (Table 9, this report).

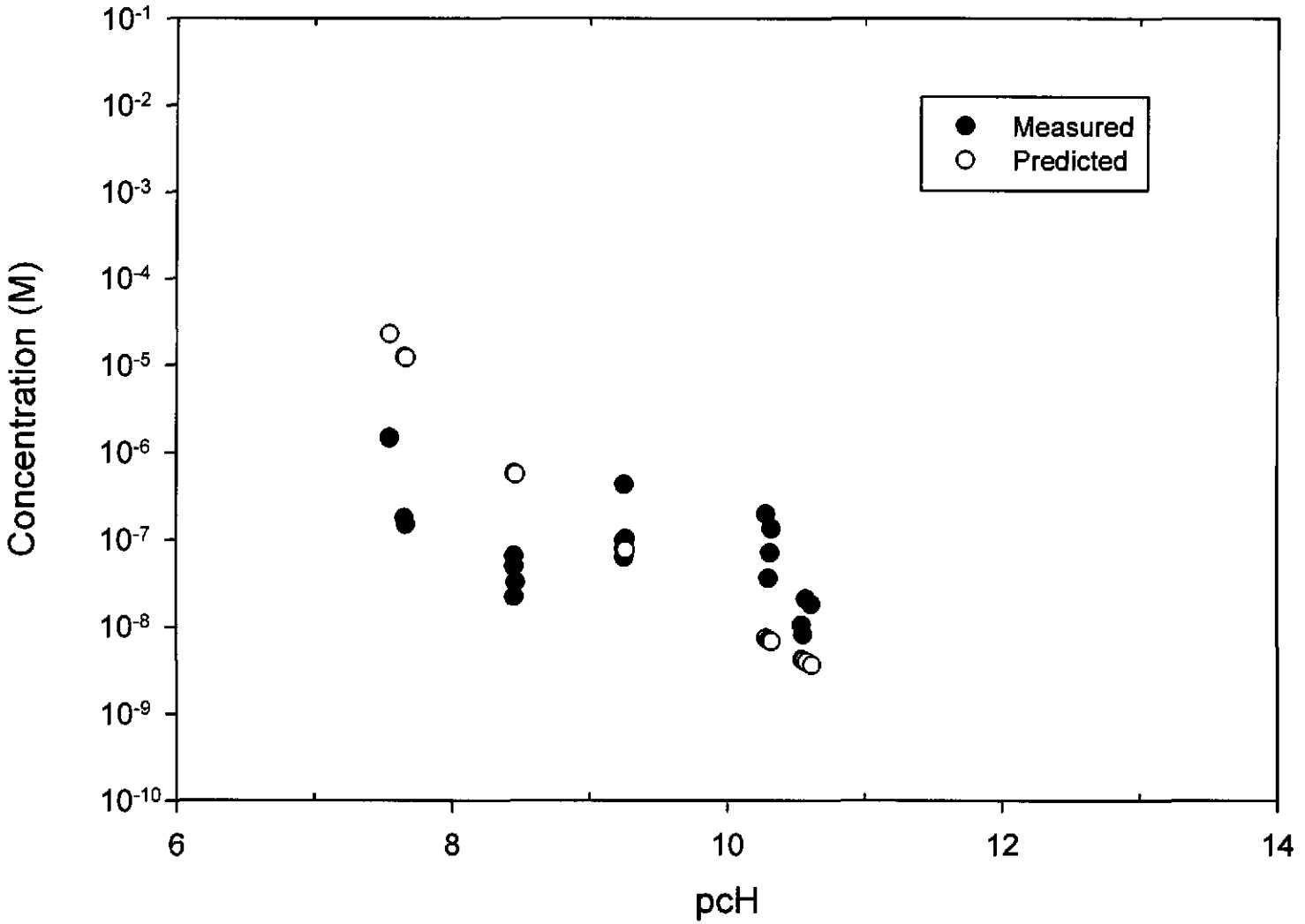

Figure 6. Comparison of the 20 Nd(III) solubilities measured in CO_3^{2-}-free ERDA-6 (Borkowski et al., 2009) with those predicted by FMT (Table 10, this report).

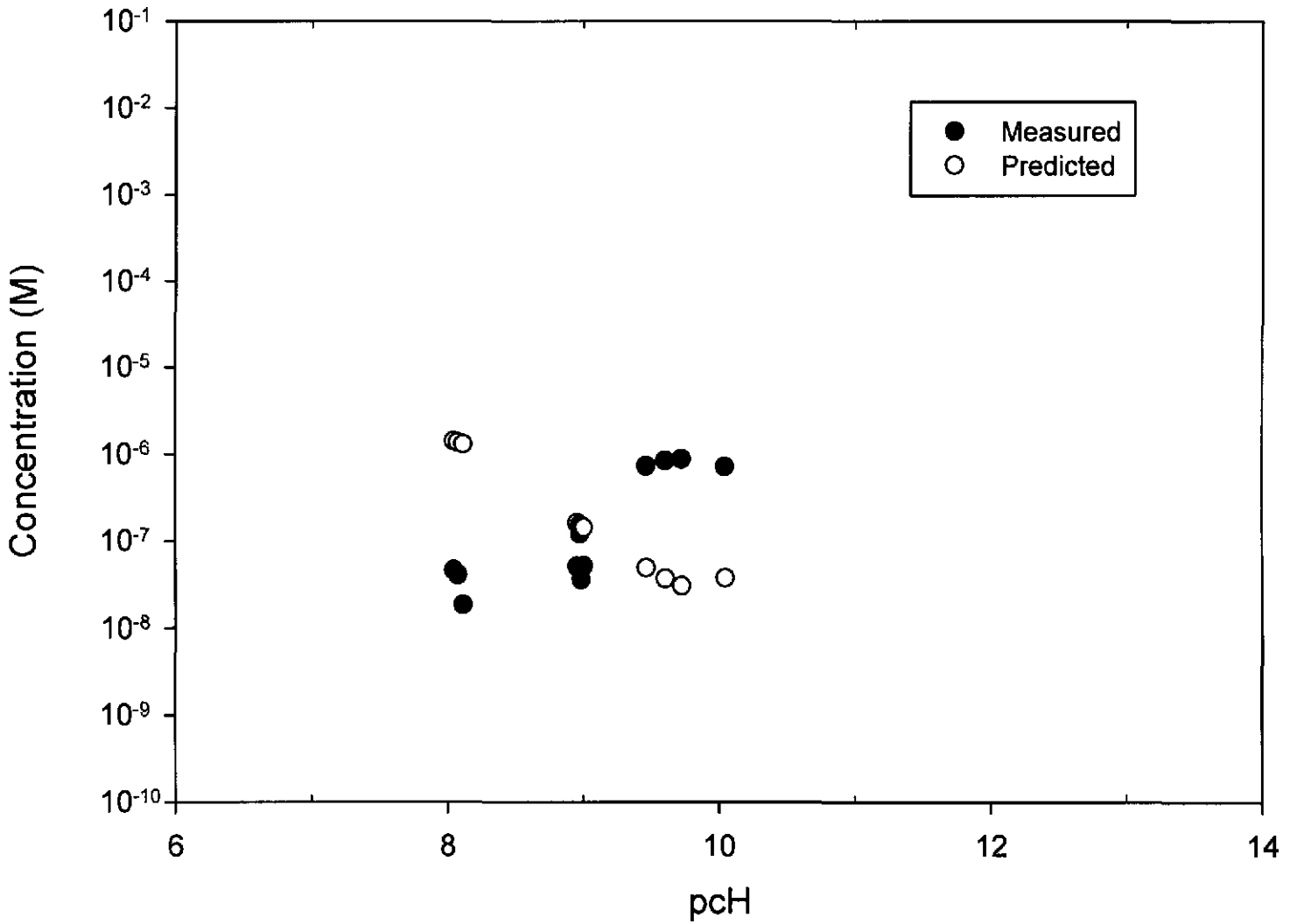

Figure 7. Comparison of the 12 Nd(III) solubilities measured in ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-5} M (Borkowski et al., 2009) with those predicted by FMT (Table 11, this report).

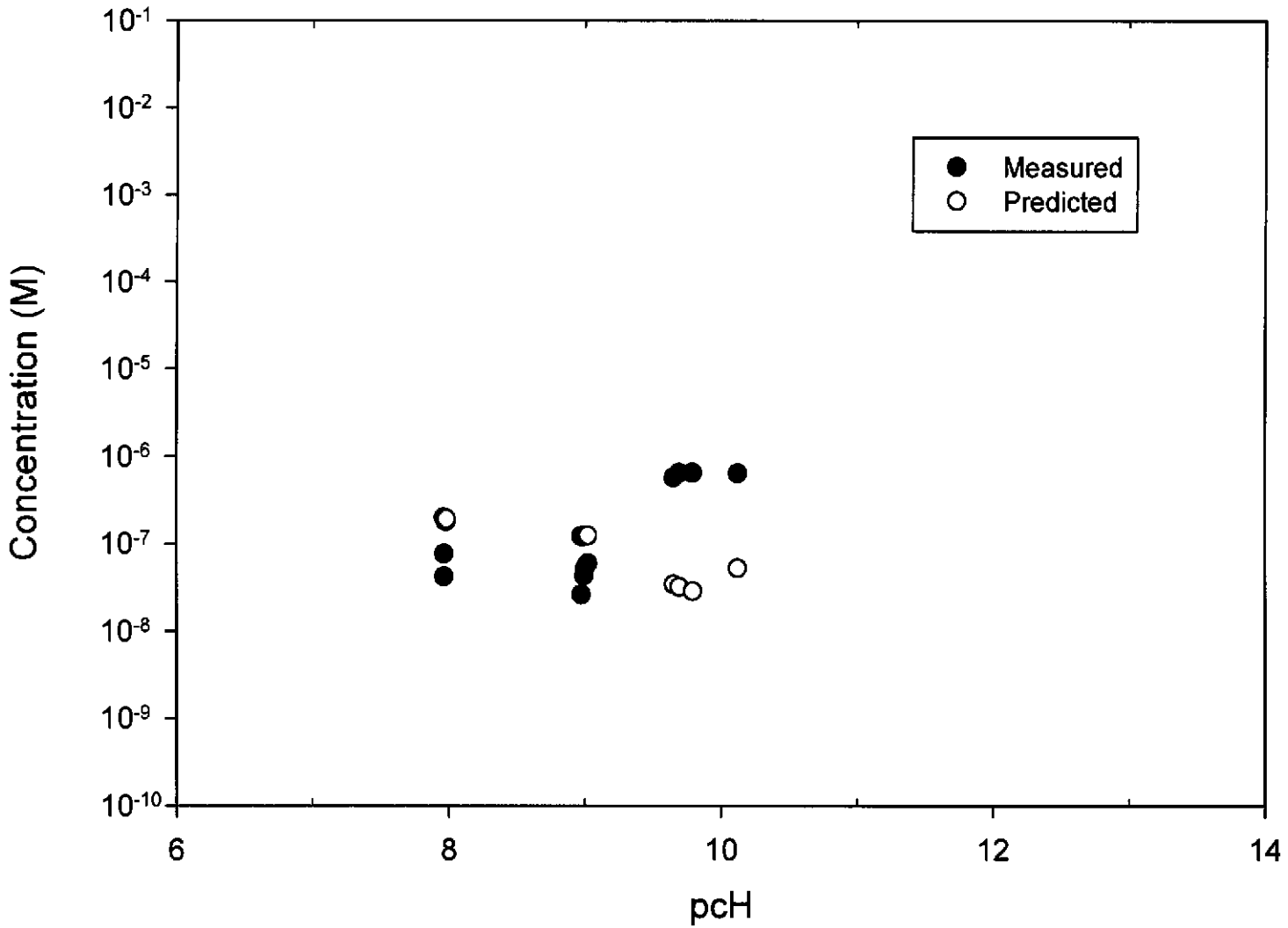

Figure 8. Comparison of the 12 Nd(III) solubilities measured in ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-4} M (Borkowski et al., 2009) with those predicted by FMT (Table 12, this report).

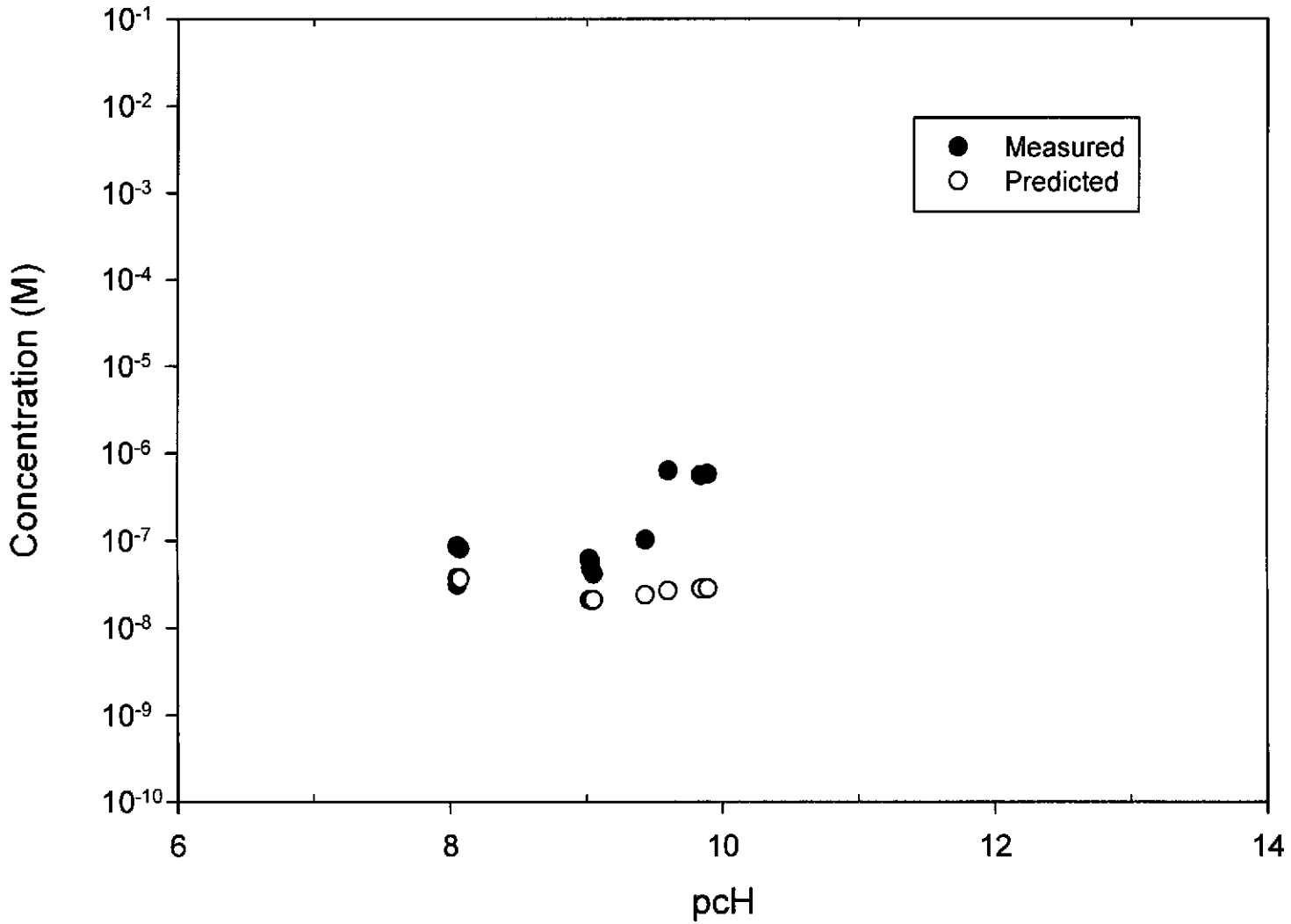

Figure 9. Comparison of the 12 Nd(III) solubilities measured in ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-3} M (Borkowski et al., 2009) with those predicted by FMT (Table 13, this report).

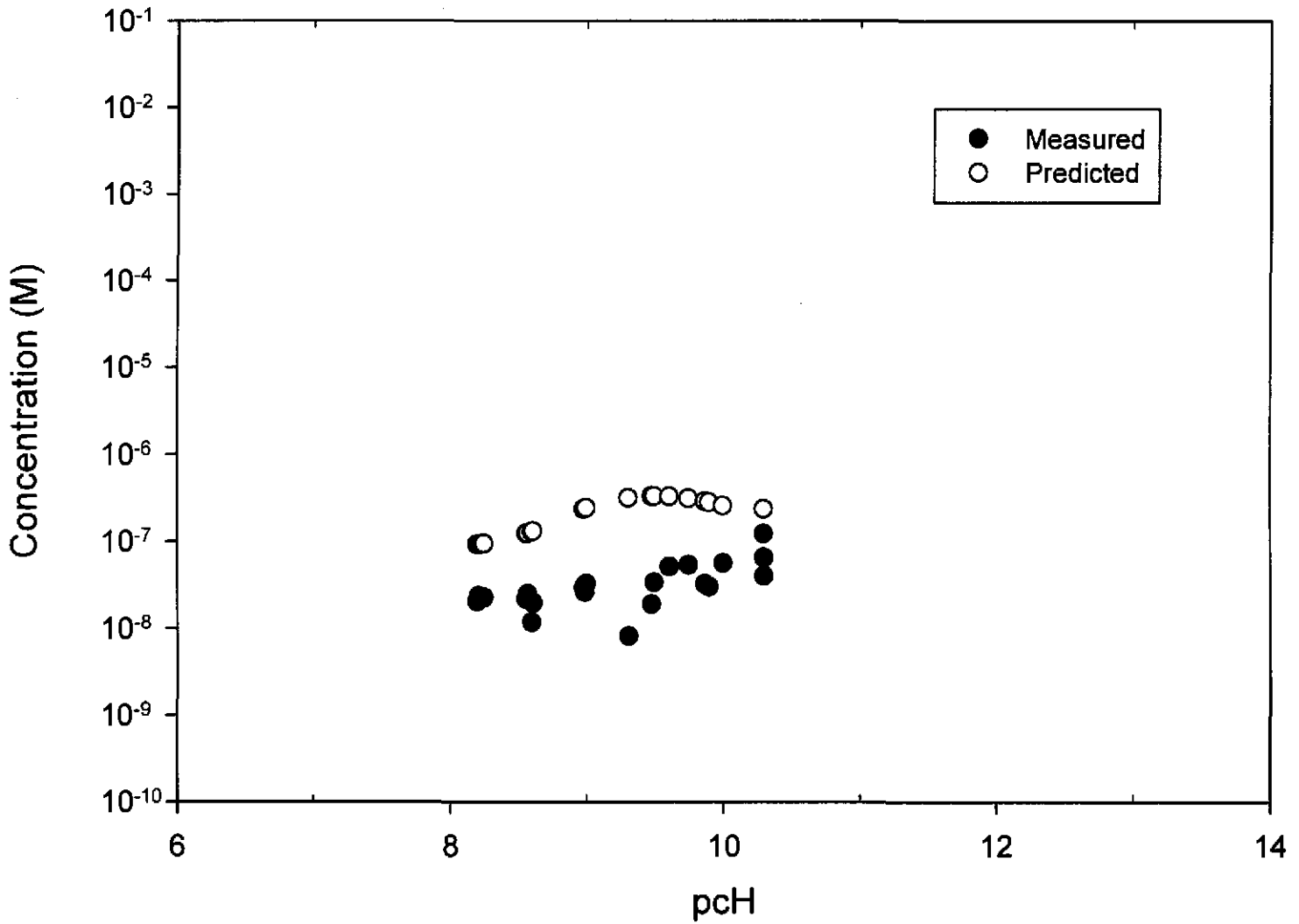

Figure 10. Comparison of the 22 Nd(III) solubilities measured in ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-2} M (Borkowski et al., 2009) with those predicted by FMT (Table 14, this report).

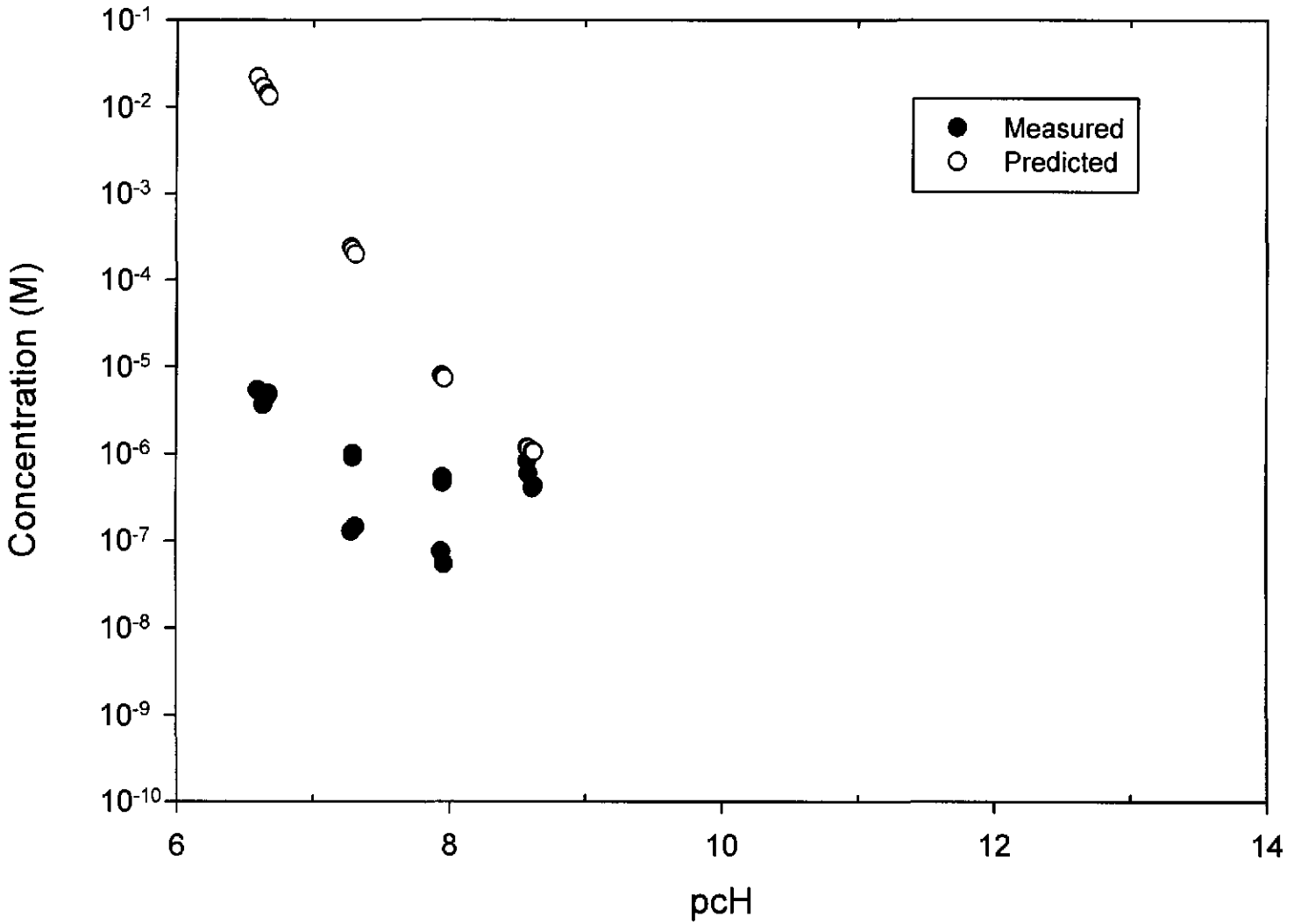

Figure 11. Comparison of the 16 Nd(III) solubilities measured in CO_3^{2-}-free GWB (Borkowski et al., 2009) with those predicted by FMT (Table 15, this report).

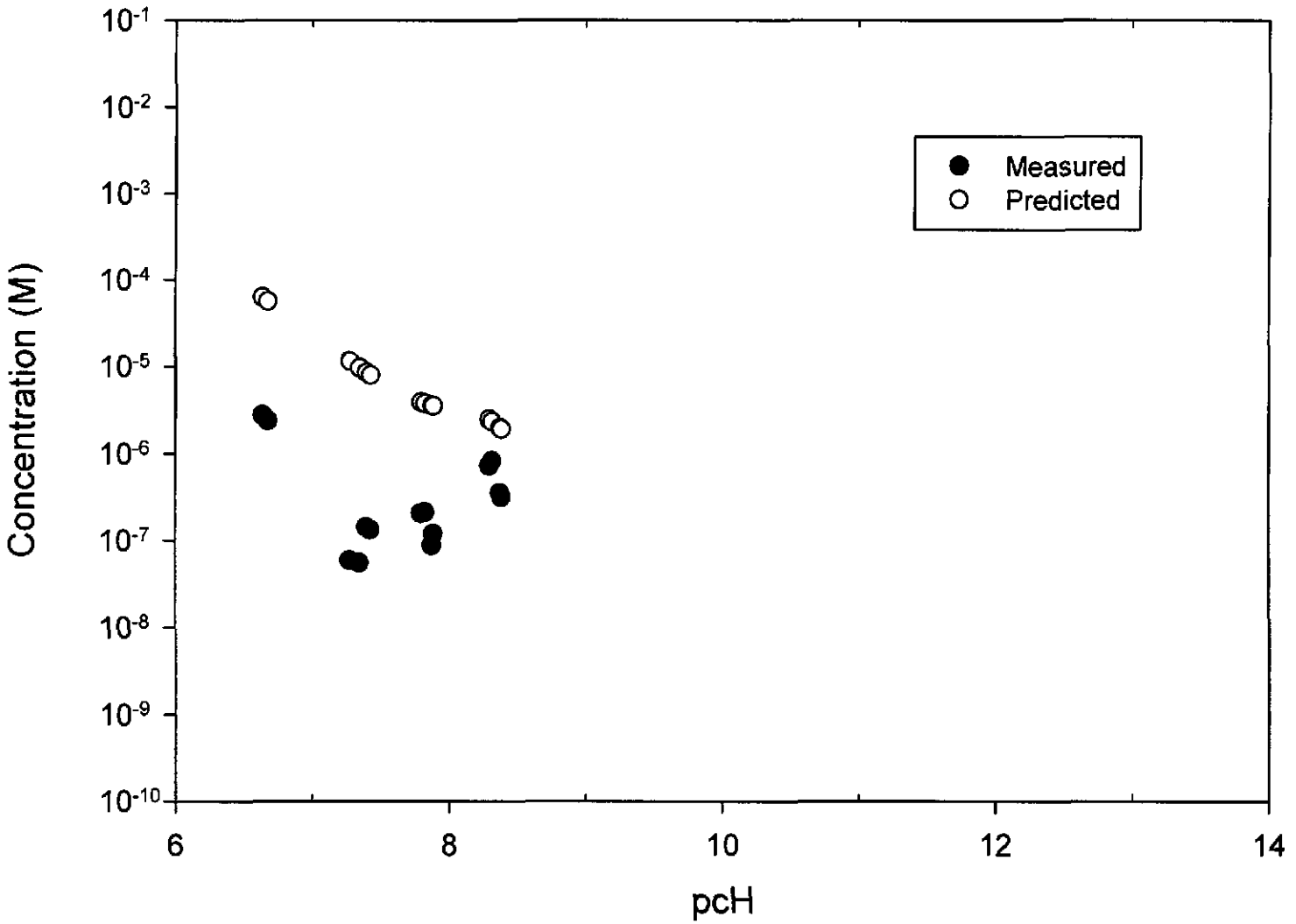

Figure 12. Comparison of the 14 Nd(III) solubilities measured in GWB with an initial CO_3^{2-} concentration of 1×10^{-5} M (Borkowski et al., 2009) with those predicted by FMT (Table 16, this report).

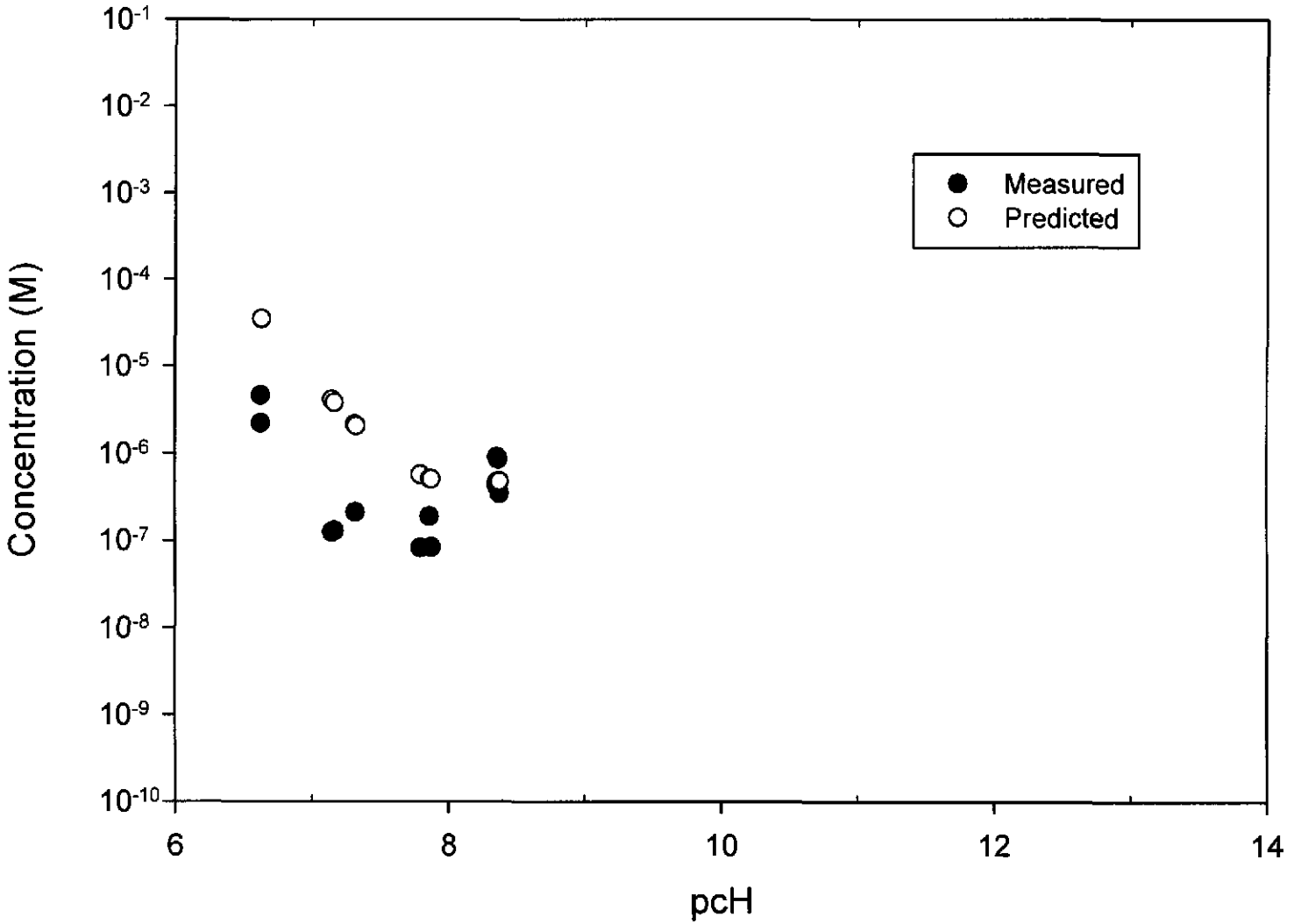

Figure 13. Comparison of the 14 Nd(III) solubilities measured in GWB with an initial CO_3^{2-} concentration of 1×10^{-4} M (Borkowski et al., 2009) with those predicted by FMT (Table 17, this report).

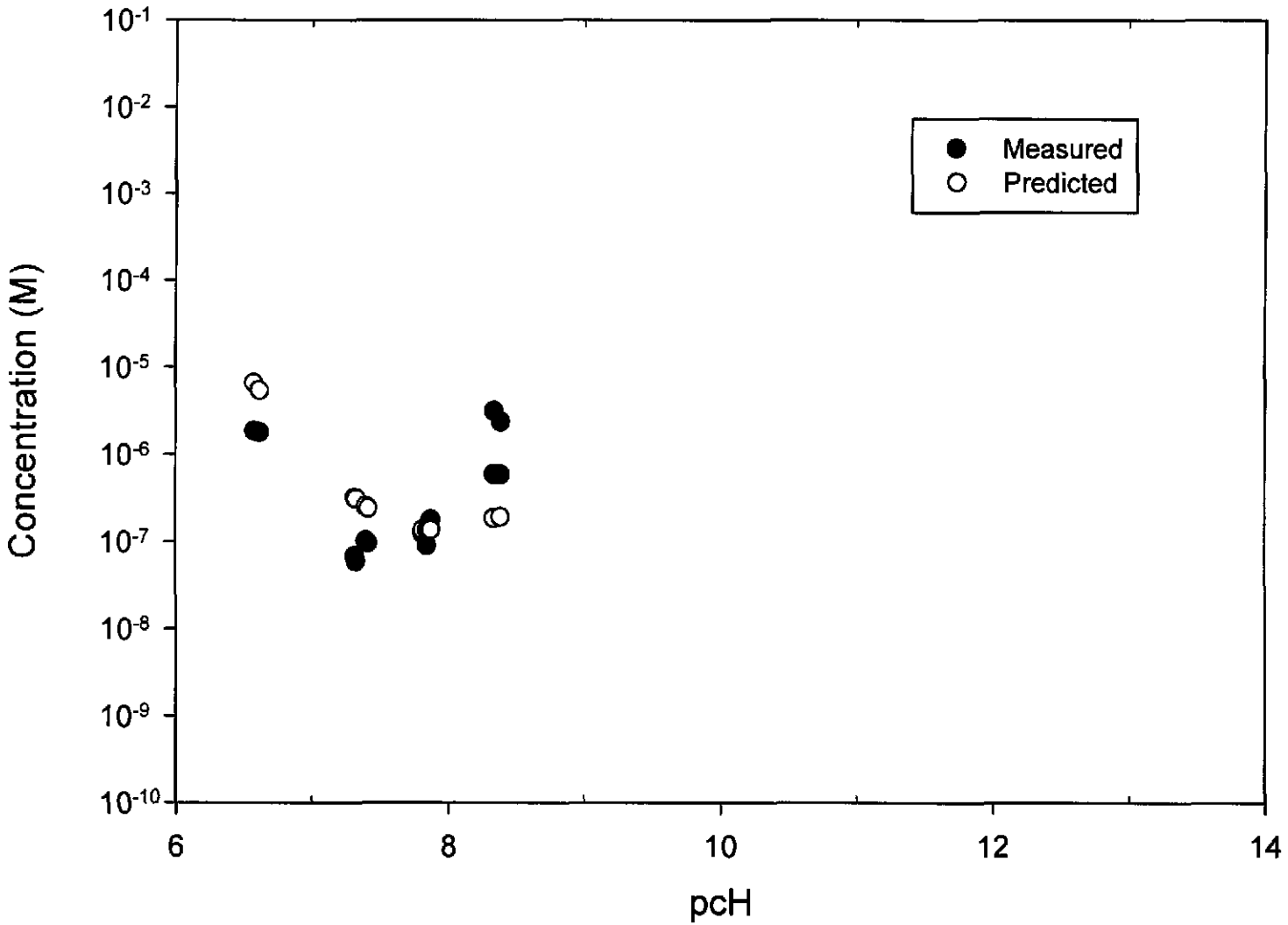

Figure 14. Comparison of the 14 Nd(III) solubilities measured in GWB with an initial CO_3^{2-} concentration of 1×10^{-3} M (Borkowski et al., 2009) with those predicted by FMT (Table 18, this report).

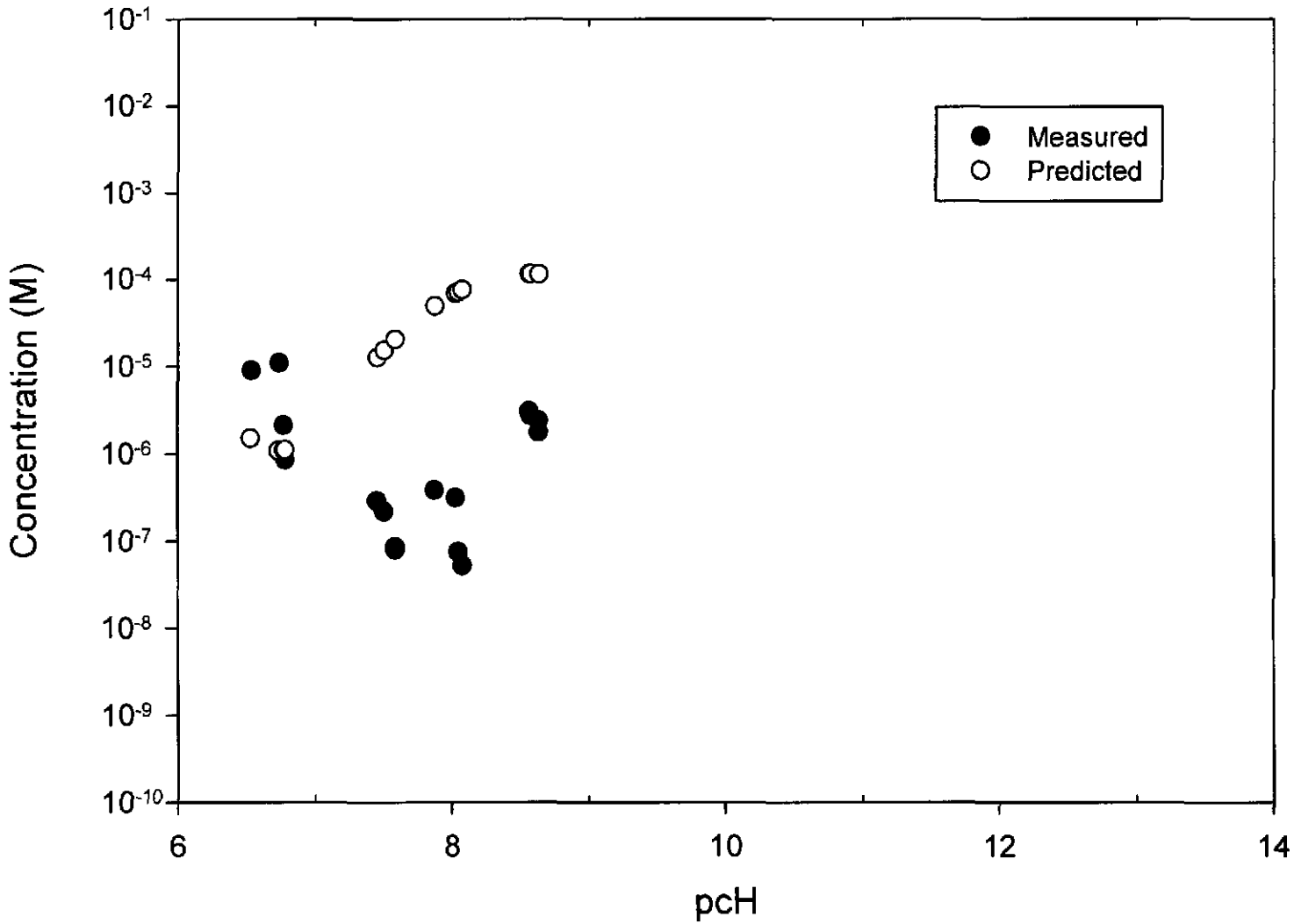

Figure 15. Comparison of the 16 Nd(III) solubilities measured in GWB with an initial CO_3^{2-} concentration of 1×10^{-2} M (Borkowski et al., 2009) with those predicted by FMT (Table 19, this report).

Table 8 and Figure 4 also show that FMT overpredicted the Nd(III) solubilities measured over the entire range of final pcH values for 5 M NaCl solutions with an initial CO_3^{2-} concentration of 1×10^{-3} M.

FMT overpredicted all of the Nd(III) solubilities measured at final pcH values ≤ 10.30; FMT underpredicted all of the solubilities at pcH ≥ 11.91 for the 5 M NaCl solutions with an initial CO_3^{2-} concentration of 1×10^{-2} M (see Table 9 and Figure 5).

3.1.2 Results for Borkowski et al. (2009), ERDA-6

Table 10 and Figure 6 illustrate that for CO_3^{2-}-free ERDA-6, FMT overpredicted the Nd(III) solubilities measured at final pcH values ≤ 9.27, with the exception of two measured solubilities at pcH = 9.26 and one at pcH = 9.27. However, FMT underpredicted all of the solubilities measured at pcH ≥ 10.29.

Table 11 and Figure 7 show that, for ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-5} M, FMT overpredicted all of the measured Nd(III) solubilities with final pcH values ≤ 9.01; and that FMT underpredicted all of the measured solubilities with pcH ≥ 9.47.

Inspection of Table 12 and Figure 8 reveals that, for ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-4} M, the results are very similar to those obtained with this brine with an initial CO_3^{2-} concentration of 1×10^{-5} M: FMT overpredicted all of the measured Nd(III) solubilities with final pcH values ≤ 9.03; and that FMT underpredicted all of the measured solubilities with pcH ≥ 9.66.

Table 13 and Figure 9 show that, for ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-3} M, FMT underpredicted the Nd(III) solubilities measured over the entire range of final pcH values, with the exception of one measured at pcH = 8.06.

For ERDA-6 with an initial CO_3^{2-} concentration of 1×10^{-2} M (Table 14 and Figure I0), FMT overpredicted the Nd(III) solubilities measured over the entire range of final pcH values.

For their calculations of the CRA-2009 PABC baseline solubilities, Brush and Xiong (2009b and 2009c, Table 9) predicted that the pcH and TIC concentration of ERDA-6 will be 9.68 and 4.48×10^{-4} M, respectively, after this brine equilibrates with the solids in WIPP disposal rooms. Inspection of Figures 8 and 9 suggests that FMT would have underpredicted these solubilities had they been measured at a pcH of 9.68 and an initial CO_3^{2-} concentration of 4.48×10^{-4} M. This is probably due to the fact that the WIPP Am(III) speciation and solubility model currently implemented in FMT does not include the Nd(III)-borate complex identified by Borkowski et al. (2009) as the dominant Nd(III)-bearing species at values of pcH close to that predicted for ERDA-6 after it equilibrates with the solids in the repository.

3.1.3 Results for Borkowski et al. (2009), GWB

Table 15 and Figure 11 indicate that, for CO_3^{2-}-free GWB, FMT overpredicted the Nd(III) solubilities measured over the entire range of final pcH values.

For GWB with an initial CO_3^{2-} concentration of 1×10^{-5} M, Table 16 and Figure 12 show results similar to those for Figure 11.

Table 17 and Figure 13 show that FMT overpredicted most of the Nd(III) solubilities measured in GWB with an initial CO_3^{2-} concentration of 1×10^{-4} M, with the exceptions of two measured at pcH = 8.36 and 8.37.

According to Table 18 and Figure 14, FMT overpredicted all of the Nd(III) solubilities measured at final pcH values ≤ 7.85; and underpredicted all of the solubilities measured at pcH $\geq$ 7.87 for GWB with an initial CO_3^{2-} concentration of 1×10^{-3} M.

Finally, for GWB with an initial CO_3^{2-} concentration of 1×10^{-2} M (Table 19 and Figure 15), FMT overpredicted most of the Nd(III) solubilities measured over the entire range of final pcH, with the exceptions of three measured at pcH = 6.54, 6.74, and 6.78.

For their calculations of the CRA-2009 PABC baseline solubilities, Brush and Xiong (2009b and 2009c, Table 8) predicted that the pcH and total inorganic C (TIC) concentration of GWB will be 9.40 and 3.50×10^{-4} M after this brine equilibrates with the solids in WIPP disposal rooms. All of the Nd(III) solubilities measured by Borkowski et al. (2009) in GWB had final pcH values ≤ 8.64. Therefore, we cannot compare their measured solubilities with those predicted for a pcH of 9.40 and a TIC concentration of 3.50×10^{-4} M.

3.2 Comment 4-C-35

Comment 4-C-35 of the EPA's February 22, 2010, letter to the DOE (Kelly, 2010) requested that "[the] DOE should examine whether the thorium concentrations predicted by FMT modeling consistently differ from the ... experimentally measured [Th] concentrations in carbonate-bearing solutions" reported by "Östhols et al. (1994), Rai et al. (1995), Felmy et al. (1997), Altmaier et al. (2005), and Altmaier et al. (2006)."

3.2.1 Results for Rai et al. (1995)

Tables 20 and 21 provide the values of the pcH calculated by FMT; the initial Na_2CO_3 and NaOH concentrations and final, measured Th(IV) concentrations (solubilities) from Rai et al. (1995, Figures 4 and 5, respectively); and the Th(IV) solubilities predicted by FMT

(this analysis) and the FMT run numbers. If two runs had the same calculated pcH, we arranged them in order of increasing initial Na_2CO_3 concentration. If two runs had the same calculated pcH and initial Na_2CO_3 concentration, we arranged them in order of increasing measured Th(IV) solubility.

Table 20 and Figure 16 show that FMT underpredicted four of the seven Th(IV) solubilities measured by Rai et al. (1995) in their Figure 4 and overpredicted the other three.

Table 21 and Figure 17 show that FMT overpredicted the Th(IV) solubilities measured by Rai et al. (1995) in their Figure 5 at NaOH $\leq 1.84 \times 10^{-2}$ m and mostly underpredicted their solubilities measured at NaOH $\geq 3.28 \times 10^{-2}$ m.

3.2.2 Results for Altmaier et al. (2005) and Altmaier et al. (2006)

Table 22 provides the values of the pcH calculated by FMT; the initial Na_2CO_3 and NaOH concentrations, and measured Th(IV) concentrations (solubilities) from Altmaier et al. (2005, Figure 4b); and the Th(IV) solubilities predicted by FMT and the FMT run numbers (this analysis) for the four experiments by Altmaier et al. (2005, Figure 4b) in which I $\geq$ 3 M. Figure 18 shows that FMT overpredicted the results of all of these measured solubilities.

Table 23 provides the final measured values of the pcH, the initial NaCl and ($NaHCO_3$-Na_2CO_3) concentrations, and the measured Th(IV) solubilities from Altmaier et al. (2006, Figure 2); and the Th(IV) solubilities predicted by FMT and the run numbers for the 12 experiments in which I $\geq$ 3 M. Figure 19 shows that FMT underpredicted all of them.

FMT underpredicted most of the solubilities measured by Altmaier et al. (2005, 2006) in experiments with I $\geq$ 3 M, probably because the WIPP Th(IV) speciation and solubility model implemented in FMT does not include the $Th(OH)_y(CO_3)_z^{4-y-2z}$ complexes suggested by Altmaier et al. (2005, 2006) as the dominant Th(IV)-bearing species in these experiments. Altmaier et al. (2005) concluded that, at high CO_3^{2-} concentrations ($\log[CO_3^{2-}]$ greater than about -0.5) in their Figure 4b, the dominant aqueous species is (are) $ThOH(CO_3)_4^{5-}$ (or $ThOH(CO_3)_4^{5-}$ and $Th(OH)_2(CO_3)_4^{6-}$), neither of which is included in the WIPP Th(IV) model. Altmaier et al., 2006, Figure 2) concluded that $ThOH(CO_3)_4^{5-}$ was the dominant Th(IV) species.

The WIPP Th(IV) model includes the following $Th(OH)_y(CO_3)_z^{4-y-2z}$ complexes: $Th(CO_3)_5^{6-}$, $Th(OH)_3CO_3^-$, and $Th(OH)_4$(aq). It does not include $ThOH(CO_3)_4^{5-}$ and $Th(OH)_2(CO_3)_2^{2-}$, both of which were suggested by Altmaier et al. (2005, Figure 5) as important at high CO_3^{2-} concentrations (see below). Furthermore, it does not include $Th(OH)_2CO_3$(aq) and $Th(OH)_4CO_3^{2-}$, which – although not identified as important in their Figure 5 – were shown by Altmaier et al. (2005) to contribute to the solubility of Th(IV) at high CO_3^{2-} concentrations.

Nevertheless, the WIPP Th(IV) model is still adequate for WIPP compliance-related calculations. This is because none of the important $Th(OH)_y(CO_3)_z^{4-y-2z}$ complexes with z > 0 approaches the $Th(OH)_4$(aq) concentrations predicted by Brush and Xiong (2009c, Tables 13 and

14) for the CRA-2009 PABC (4.52×10^{-8} M for GWB and 4.76×10^{-8} M for ERDA-6 until the log of the CO_3^{2-} concentration exceeds about −1 (see Altmaier et al., 2005, Figure 4b). By contrast, the TIC concentrations (essentially the sum of the concentrations of HCO_3^- and CO_3^{2-}) predicted by Brush and Xiong (2009c, Tables 8 and 9), 0.350 mM for GWB and 0.448 mM for ERDA-6, were more than two orders of magnitude lower than that threshold. The reason why the TIC concentrations predicted for these WIPP brines are too low to form important $Th(OH)_y(CO_3)_z^{4-y-2z}$ complexes with $z > 0$ is because the brucite-hydromagnesite carbonation reaction will buffer the f_{CO_2} at 3.14×10^{-6} atm in both GWB and ERDA-6. This value of f_{CO_2} will in turn maintain the TIC at 0.350 mM in GWB and 0.448 mM in ERDA-6.

3.3 Comment 4-C-36

EPA Comment 4-C-36 requests that, in view of the results reported by Altmaier et al. (2004), the DOE address (1) whether significant concentrations of Th(IV) intrinsic colloids (eigencolloids) or mineral-fragment colloids (pseudocolloids) could form in the WIPP, and (2) if so, what would be the effects of such colloids on PA.

Altmaier et al. (2004) measured the solubilities of ThO_2(cr) and $ThO_n(OH)_{4-2n} \cdot xH_2O$(am) in carbonate-free 0.5 and 5 M NaCl solutions and 0.25, 2.5, and 4.5 M $MgCl_2$ solutions. They observed intrinsic colloids in their experiments in 0.5 and 5 M NaCl, and mineral-fragment colloids in 2.5 and 4.5 M $MgCl_2$. They measured the concentrations of Th(IV) dissolved species (mainly $Th(OH)_4$(aq)) and Th(IV) intrinsic colloids (mainly $Th(OH)_4$(col)) by comparing the Th(IV) concentrations obtained from uncentrifuged, unfiltered samples and ultracentrifuged samples (rotation velocities ≥ 50,000 rpm); they measured the concentrations of mineral-fragment colloids, which they identified as phase 3 ($Mg_2(OH)_3Cl \cdot 4H_2O$), by comparing results obtained from uncentrifuged, unfiltered samples and ultrafiltered samples (2 nm filtration).

Xiong et al. (2009) included all six of the uncentrifuged results from solutions with $I \geq 3$ M from Altmaier et al. (2004, Figure 2) in their Th(IV) uncertainty analysis: two from their 5 M NaCl solutions, two from 2.5 M $MgCl_2$ solutions, and two from 4.5 M $MgCl_2$ solutions. However, they excluded the ultracentrifuged results of Altmaier et al. (2004) because this posttest phase-separation method was significantly different from those employed in the studies used to parameterize the WIPP Th(IV) solubility model (e.g., centrifugation or filtration) and appeared to support a significantly different solubility model than that established for the WIPP (e.g., ultracentrifugation showed that 99% or more of what had been interpreted as dissolved Th(IV) present as the $Th(OH)_4$(aq) species appeared to be present as the intrinsic colloid $Th(OH)_4$(col)). The WIPP Th(IV) model was parameterized in the mid-1990s, before it was realized that the phase-separation methods used then might not have removed all of the $Th(OH)_4$(col). Thus, Xiong et al. (2009) excluded the ultracentrifuged results of Altmaier et al. (2004) to be consistent with the WIPP Th(IV) model.

This subsection (see below) explains why the types and concentrations of colloids reported by Altmaier et al. (2004) do not appear to be relevant to the WIPP. Because we concluded that these colloids will not form in the WIPP, we did not attempt to assess the effects of these colloids on PA.

Table 20. Comparison of the Seven Th(IV) Solubilities Measured in Solutions with I ≥ 3 M (Rai et al., 1995, Figure 4) with Those Predicted by FMT (This Analysis). See Figure 16 (this report) for the scatter plot of measured and predicted solubilities.

Calculated pC_H	Initial Na_2CO_3 Conc. (m)	Initial NaOH Conc. (M)	Th(IV) Solubility, Measured (m)	Th(IV) Solubility, Predicted (m)	FMT Run Number
12.36	9.8×10^{-1}	1×10^{-1}	8.87×10^{-7}	5.77×10^{-7}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_001
12.36	9.8×10^{-1}	1×10^{-1}	6.16×10^{-7}	5.77×10^{-7}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_001
12.36	9.8×10^{-1}	1×10^{-1}	3.46×10^{-6}	5.77×10^{-7}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_001
12.45	1.48×10^{0}	1×10^{-1}	3.38×10^{-6}	1.42×10^{-5}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_003
12.45	1.48×10^{0}	1×10^{-1}	1.82×10^{-5}	1.42×10^{-5}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_003
12.52	1.95×10^{0}	1×10^{-1}	1.10×10^{-5}	1.84×10^{-4}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_005
12.52	1.95×10^{0}	1×10^{-1}	6.68×10^{-5}	1.84×10^{-4}	FMT_PABC09_RAI95_1E-1M_NAOH_NA2CO3_005

Table 21. Comparison of the 19 Th(IV) Solubilities Measured in Solutions with I ≥ 3 M (Rai et al., 1995, Figure 5) with Those Predicted by FMT (This Analysis). See Figure 17 (this report) for the scatter plot of measured and predicted solubilities.

Calculated pC_H	Initial Na_2CO_3 Conc. (M)	Initial NaOH Conc. (m)	Th(IV) Solubility, Measured (m)	Th(IV) Solubility, Predicted (m)	FMT Run Number
11.57	1.0×10^{0}	9.86×10^{-3}	2.75×10^{-4}	5.22×10^{-4}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_014
11.58	1.0×10^{0}	9.91×10^{-3}	2.16×10^{-4}	5.18×10^{-4}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_015
11.59	1.0×10^{0}	1.07×10^{-2}	1.56×10^{-4}	4.59×10^{-4}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_006
11.73	1.0×10^{0}	1.84×10^{-2}	8.33×10^{-5}	1.42×10^{-4}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_007
11.73	1.0×10^{0}	1.84×10^{-2}	9.60×10^{-5}	1.42×10^{-4}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_007
11.94	1.0×10^{0}	3.28×10^{-2}	4.27×10^{-5}	2.28×10^{-5}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_008
12.06	1.0×10^{0}	4.37×10^{-2}	1.83×10^{-5}	8.48×10^{-6}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_009
12.11	1.0×10^{0}	4.96×10^{-2}	5.03×10^{-6}	5.48×10^{-6}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_010
12.11	1.0×10^{0}	4.96×10^{-2}	6.28×10^{-6}	5.48×10^{-6}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_010
12.11	1.0×10^{0}	4.99×10^{-2}	2.42×10^{-5}	5.40×10^{-6}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_011
12.40	1.0×10^{0}	9.81×10^{-2}	2.33×10^{-6}	6.77×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_012
12.40	1.0×10^{0}	9.86×10^{-2}	8.33×10^{-7}	6.69×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_013
12.40	1.0×10^{0}	9.86×10^{-2}	1.15×10^{-6}	6.69×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_013
12.90	1.0×10^{0}	2.95×10^{-1}	7.18×10^{-7}	1.44×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_001
12.90	1.0×10^{0}	2.96×10^{-1}	3.55×10^{-7}	1.44×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_002
12.91	1.0×10^{0}	3.00×10^{-1}	2.62×10^{-7}	1.43×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_003
13.14	1.0×10^{0}	4.89×10^{-1}	7.20×10^{-8}	1.15×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_004
13.14	1.0×10^{0}	4.89×10^{-1}	1.89×10^{-7}	1.15×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_004
13.14	1.0×10^{0}	4.92×10^{-1}	1.10×10^{-7}	1.15×10^{-7}	FMT_PABC09_RAI95_NAOH_1M_NA2CO3_005

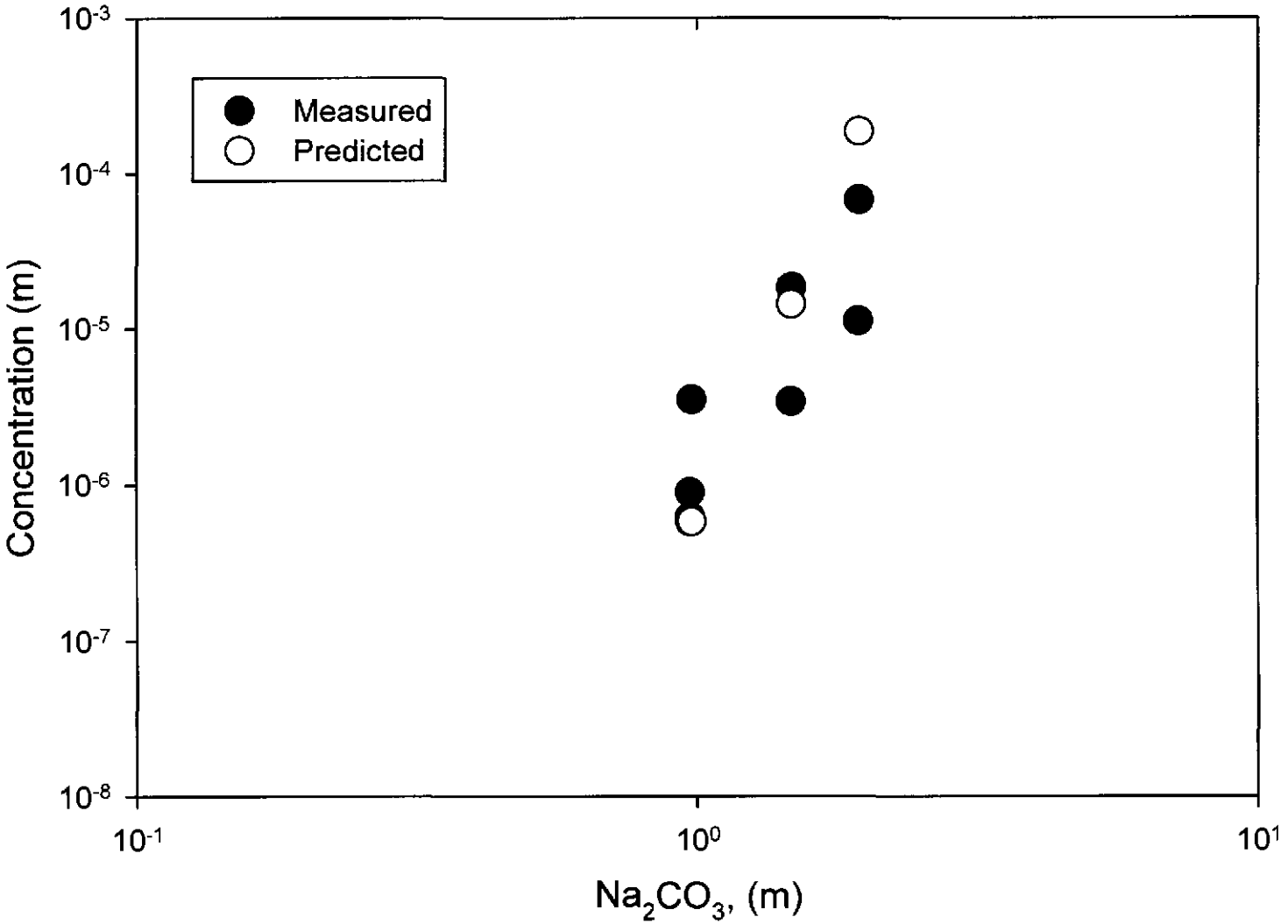

Figure 16. Comparison of the seven Th(IV) solubilities measured in solutions with $I \geq 3$ M (Rai et al., 1995, Figure 4) with those predicted by FMT (this analysis). The solutions used in these runs contained 1×10^{-1} M NaOH had calculated pcH values of 12.36 to 12.52 (Table 20, this report).

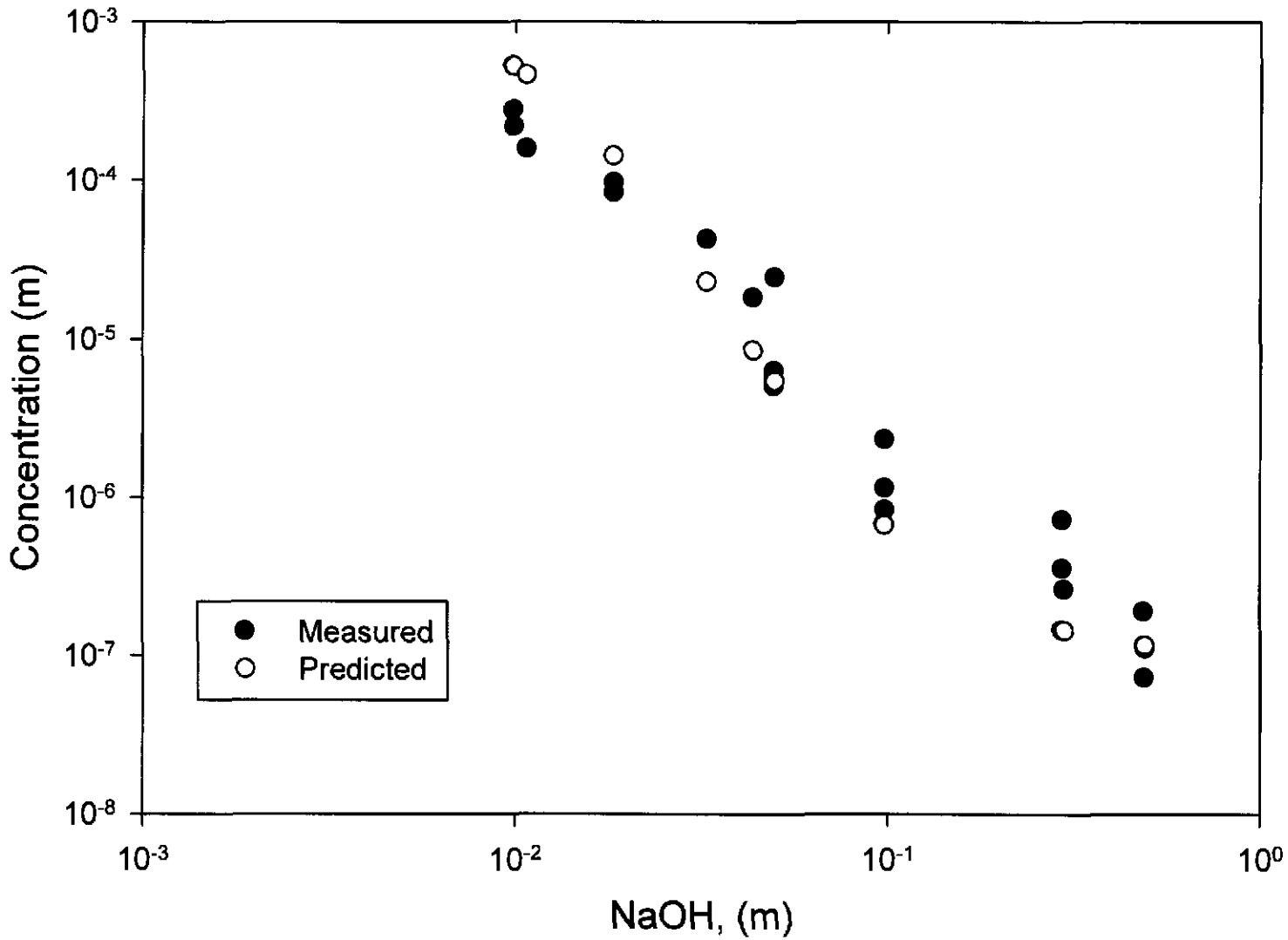

Figure 17. Comparison of the 19 Th(IV) solubilities measured in solutions with $I \geq 3$ M (Rai et al., 1995, Figure 5) with those predicted by FMT (this analysis). The solutions used in these runs contained 1.0×10^0 M Na_2CO_3 and had calculated pcH values of 11.57 to 13.14 (Table 21, this report).

Table 22. Comparison of the Four Th(IV) Solubilities Measured in Solutions with I ≥ 3 M (Altmaier et al., 2005, Figure 4b) with Those Predicted by FMT (this analysis). See Figure 18 (this report) for the scatter plot of measured and predicted solubilities.

Calculated pC_H	Initial Na_2CO_3 Conc. (M)	Initial NaOH Conc. (M or m?)[A]	Th(IV) Solubility, Measured (M)	Th(IV) Solubility, Predicted (M)	FMT Run Number
12.44	1.35×10^{0}	1×10^{-1}	3.06×10^{-6}	9.33×10^{-6}	FMT_PABC09_ALTMAIER05IV_004
12.49	1.38×10^{0}	1×10^{-1}	4.84×10^{-6}	8.18×10^{-6}	FMT_PABC09_ALTMAIER05IV_002
12.51	1.78×10^{0}	1×10^{-1}	1.48×10^{-5}	1.42×10^{-4}	FMT_PABC09_ALTMAIER05IV_003
12.52	1.82×10^{0}	1×10^{-1}	1.75×10^{-5}	1.75×10^{-4}	FMT_PABC09_ALTMAIER05IV_001

A. Altmaier et al. (2005, Figure 4b, state that the concentration of NaOH in these experiments was both 1×10^{-1} M and 1×10^{-1} m. We do not know which concentration they actually used.

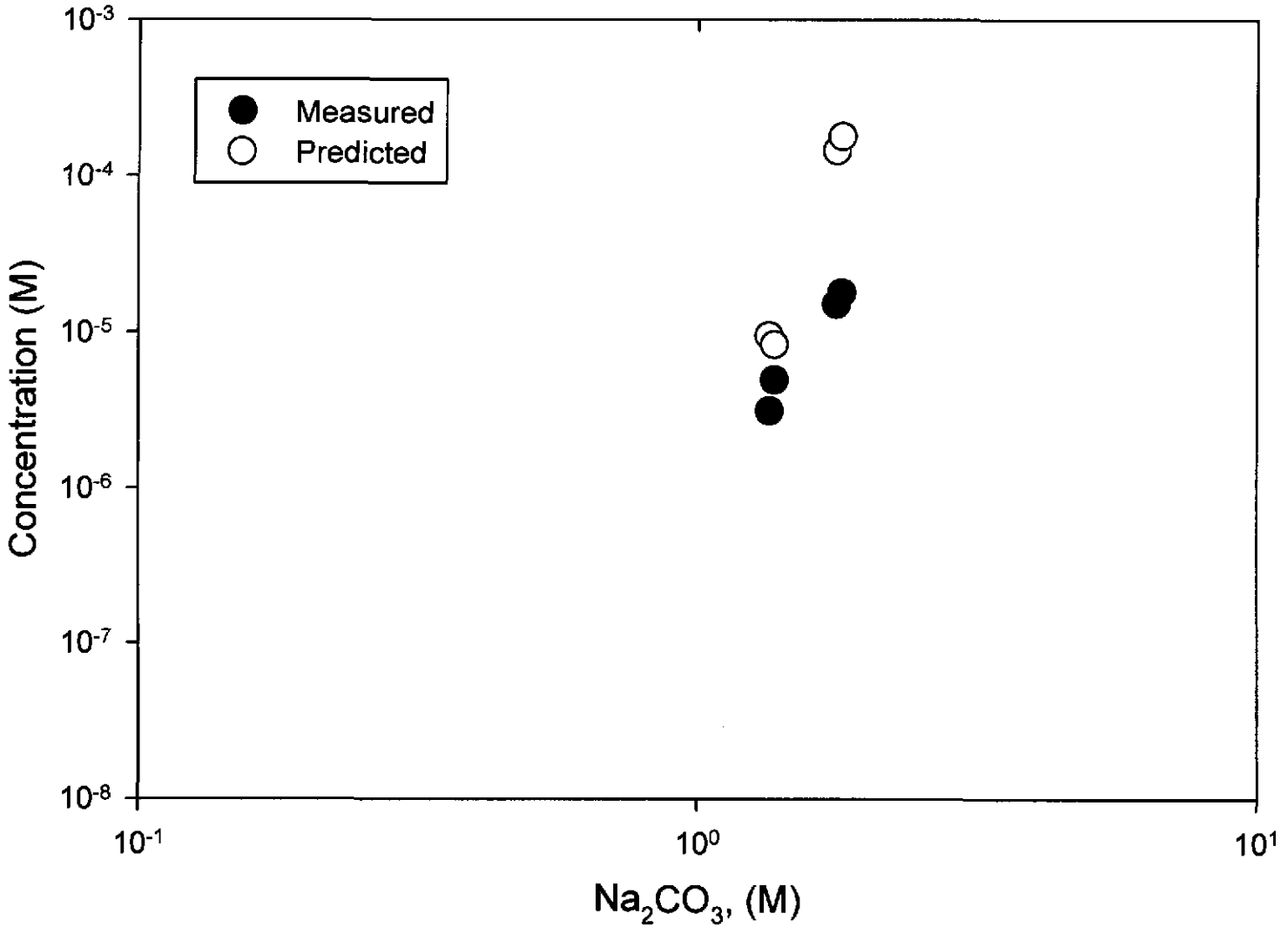

Figure 18. Comparison of the four Th(IV) solubilities measured in solutions with $I \geq 3$ M (Altmaier et al., 2005, Figure 4b) with those predicted by FMT (this analysis). The solutions used in these runs contained 1×10^{-1} M NaOH and had calculated pcH values of 12.44 to 12.52 (Table 22, this report).

Table 23. Comparison of the 12 Th(IV) Solubilities Measured in Solutions with I ≥ 3 M (Altmaier et al., 2006, Figure 2) with Those Predicted by FMT (this analysis). See Figure 19 (this report) for the scatter plot of measured and predicted solubilities.

Measured pC_H	Initial NaCl Conc. (M)	Initial $NaHCO_3$ + Na_2CO_3 Conc. (M)	Th(IV) Solubility, Measured (M)	Th(IV) Solubility, Predicted (M)	FMT Run Number
8.61	3.98×10^{0}	2.00×10^{-2}	2.64×10^{-4}	2.90×10^{-5}	FMT_PABC09_ALTMAIER06IV_001
8.65	3.98×10^{0}	2.00×10^{-2}	3.11×10^{-4}	3.02×10^{-5}	FMT_PABC09_ALTMAIER06IV_002
8.78	3.98×10^{0}	2.00×10^{-2}	3.67×10^{-4}	3.38×10^{-5}	FMT_PABC09_ALTMAIER06IV_003
8.80	3.98×10^{0}	2.00×10^{-2}	3.80×10^{-4}	3.42×10^{-5}	FMT_PABC09_ALTMAIER06IV_004
8.97	3.98×10^{0}	2.00×10^{-2}	3.79×10^{-4}	3.70×10^{-5}	FMT_PABC09_ALTMAIER06IV_005
9.00	3.98×10^{0}	2.00×10^{-2}	4.05×10^{-4}	3.72×10^{-5}	FMT_PABC09_ALTMAIER06IV_006
9.25	3.98×10^{0}	2.00×10^{-2}	3.76×10^{-4}	3.39×10^{-5}	FMT_PABC09_ALTMAIER06IV_007
9.26	3.98×10^{0}	2.00×10^{-2}	4.16×10^{-4}	3.36×10^{-5}	FMT_PABC09_ALTMAIER06IV_008
9.63	3.98×10^{0}	2.00×10^{-2}	3.99×10^{-4}	1.65×10^{-5}	FMT_PABC09_ALTMAIER06IV_009
9.64	3.98×10^{0}	2.00×10^{-2}	4.57×10^{-4}	1.61×10^{-5}	FMT_PABC09_ALTMAIER06IV_010
9.85	3.98×10^{0}	2.00×10^{-2}	2.57×10^{-4}	7.88×10^{-6}	FMT_PABC09_ALTMAIER06IV_011
9.86	3.98×10^{0}	2.00×10^{-2}	2.94×10^{-4}	7.58×10^{-6}	FMT_PABC09_ALTMAIER06IV_012

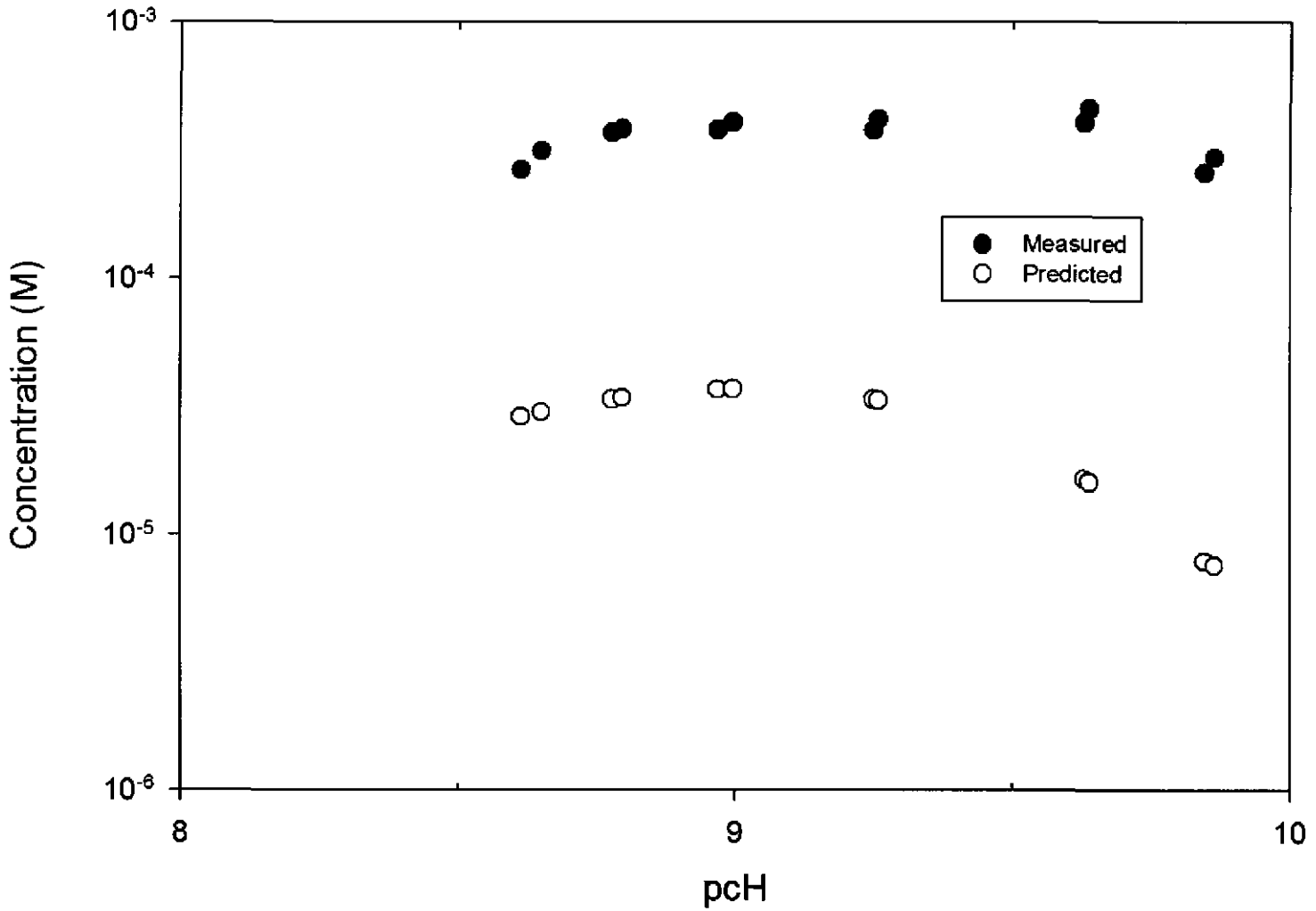

Figure 19. Comparison of the I2 Th(IV) solubilities measured in solutions with I ≥ 3 M (Altmaier et al., 2006, Figure 2) with those predicted by FMT (this analysis). The solutions used in these runs contained 3.98 M NaCl and 0.02 M ($NaHCO_3$ + Na_2CO_3), and had calculated pcH values of 8.61 to 9.86 (Table 23, this report).

There are at least five reasons why the results of Altmaier et al. (2004) are not applicable to the WIPP.

First, Altmaier et al. (2004) used pure NaCl or $MgCl_2$ solutions for their experiments, not WIPP brines such as GWB or ERDA-6. We do not know of any experiments that have identified +IV actinide intrinsic or mineral-fragment colloids in these or any other WIPP brines. In fact, the WIPP colloidal actinide source term program, carried out to support the WIPP CCA, did not identify intrinsic or mineral-fragment colloids (U.S. DOE, 1996, Appendix SOTERM, Section SOTERM.6).

Second, Altmaier et al. (2004, Subsection 3.3, p. 542) concluded that, with respect to the mineral-fragment colloids:

> The high [Th(IV)] concentrations from these pseudocolloids observed in the present laboratory experiments have not to be expected in real systems (Q-brine/brucite/magnesium hydroxychloride [phase 3]). The ratio of Th(IV) sorbed onto colloidal (mobile) and solid (immobile) magnesium hydroxychloride depends on the ratio of solution volume (saturated with $Mg_2(OH)_3Cl{\cdot}4H_2O$ colloids) and solid magnesium hydroxychloride. In our experiments, for practical reasons only 20–40 mg of the solid phase was added to 50 ml solution, whereas real systems contain large amounts of solid magnesium hydroxide/ hydroxychloride (Sorel cement) and small volumes of aqueous phase. In NaCl or dilute $MgCl_2$ solutions, neither solid $Mg_2(OH)_3Cl{\cdot}4H_2O$(s) nor its colloids are stable.

Altmaier et al. (2004, Section 4, second subsection, p. 542) added that:

> The high solubility observed in 4.5 M $MgCl_2$ due to the formation of pseudocolloids $Th(IV){\cdot}Mg_2(OH)_3Cl{\cdot}4H_2O$ (coll) has not to be expected in real systems where, contrary to the present laboratory experiments, the ratio of solution volume and solid magnesium hydroxide/hydroxychloride is very small so that sorption of Th(IV) onto solid (immobile) magnesium hydroxychloride will prevail over sorption onto (mobile) colloids. In this context it must also be emphasized that An(IV) eigencolloids, which cause relatively high solubilities, also show a high tendency towards sorption onto glass and mineral surfaces.

Third, the concentrations of Th(IV) mineral-fragment colloids, which exceeded those of the intrinsic colloids in the experiments of Altmaier et al. (2004), were proportional to the dissolved Mg concentration used in their experiments. Table 2 of Altmaier et al. (2004) shows that, in 4.5 M $MgCl_2$, the reported values of $\log[Th]_{tot}$ (the log of the concentration of mineral-fragment and intrinsic colloids plus dissolved species) were −4.7 and −4.7. These concentrations were significantly higher than the values reported for 2.5 M $MgCl_2$, −5.9 and −5.7. Most of the Th(IV) species in these uncentrifuged, unfiltered samples were mineral-fragment colloids, because: (1) the values of log[Th] (the Th(IV) solubilities) reported by Altmaier et al. (2004, Tables 2 and 3) varied from −9.1 to −7.8, and (2) there is no reason to suspect that the values of $\log[Th]_{intrinsic\ col.}$ for 2.5 and 4.5 M $MgCl_2$ would differ significantly

from those reported by Altmaier et al. (2004, Table 2) for 5 M NaCl, $\log[Th]_{intrinsic\ col.} \approx \log[Th]_{tot} = -6.3$ and -6.5.

After reaction with the solids in WIPP disposal rooms, the Mg concentration of GWB will be 0.463 M (Brush and Xiong, 2009c, Table 8). Therefore, the Mg concentrations in the 2.5 and 4.5 M $MgCl_2$ experiments by Altmaier et al. (2004), which produced Th(IV) mineral-fragment colloids, were over five and nine times higher than that expected in the repository. Consequently, the concentrations of Th(IV) mineral-fragment colloids could be much lower in the WIPP than those observed by Altmaier et al. (2004), if they form at all.

Fourth, phase 3 formed in the experiments that Altmaier et al. (2004) carried out with 2.5 and 4.5 M $MgCl_2$ solutions; this phase produced the mineral-fragment colloids that they observed. In the WIPP, however, we expect that phase 5 ($Mg_3(OH)_5Cl{\cdot}4H_2O$) will form. We expect phase 5 instead of phase 3 because: (1) phase 5 has always been observed in laboratory experiments with MgO and GWB at SNL, and (2) in calculations with FMT_090720.CHEMDAT and GWB (Brush and Xiong, 2009c), the thermodynamic database that includes both of phase 3 and phase 5, FMT has always predicted that phase 5 is stable with respect to phase 3. We do not know of any experiments that have produced mineral-fragment colloids from phase 5.

Finally, the phase 3 used by Altmaier et al. (2004) was a fine-grained powder. Therefore, when phase 3 was in contact with their solutions, dissolution and reprecipitation probably occurred. During this dissolution and reprecipitation, Th(IV) may have sorbed onto the phase 3 colloids. Under the expected WIPP conditions used in the SNL study of MgO, the reaction path is that MgO partially converts to phase 5 in GWB (Xiong and Lord, 2008), and the phase 5 produced is a well indurated (cemented) form of Mg hydroxychloride.

Therefore, the DOE does not expect the colloids observed by Altmaier et al. (2004) to form in the WIPP.

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4 CONCLUSIONS

Comparisons of the Nd(III) solubilities measured by Borkowski et al. (2009) with those predicted by FMT (this analysis) demonstrate that, for 5 M NaCl solutions, (1) FMT overpredicted or mostly overpredicted the solubilities measured at low values of final pcH, but underpredicted them at high pcH in CO_3^{2-}-free experiments and experiments with an initial CO_3^{2-} concentration of 1×10^{-2} M; and (2) FMT overpredicted or mostly overpredicted the solubilities measured in runs with initial CO_3^{2-} concentrations of 1×10^{-5} M, 1×10^{-4} M, and 1×10^{-3} M. (Subsection 3.1.1 describes these conclusions in detail.)

Analogous comparisons for ERDA-6 show that (1) FMT overpredicted the solubilities measured by Borkowski et al. (2009) at low values of final pcH, but underpredicted or mostly underpredicted them at high pcH in CO_3^{2-}-free experiments and experiments with initial CO_3^{2-} concentrations of 1×10^{-5} and $I \times 10^{-4}$ M; (2) FMT mostly underpredicted the solubilities measured in runs with an initial CO_3^{2-} concentration of 1×10^{-3} M; and (3) FMT overpredicted the solubilities measured in runs with an initial CO_3^{2-} concentration of 1×10^{-2} M. FMT underpredicted the Nd(III) solubilities measured under the conditions closest to those expected in WIPP disposal rooms (see Subsection 3.1.2).

For GWB, (1) FMT overpredicted or mostly overpredicted the solubilities measured by Borkowski et al. (2009) in CO_3^{2-}-free experiments and runs with initial CO_3^{2-} concentrations of 1×10^{-5}, 1×10^{-4}, and 1×10^{-2} M; and (2) FMT overpredicted the solubilities measured at low values of final pcH, but underpredicted them at high pcH in experiments with an initial CO_3^{2-} concentration of 1×10^{-3} M. We cannot compare solubilities measured and predicted for GWB under the conditions expected in the repository because Borkowski et al. (2009) did not carry our any experiments at the pcH predicted for GWB in the repository (Subsection 3.1.3).

Comparisons of the Nd(III) solubilities measured by Rai et al. (1995, Figure 4) with those predicted by FMT (this analysis) demonstrate that FMT underpredicted the Th(IV) solubilities measured at low Na_2CO_3 concentrations and overpredicted those at high concentrations. FMT overpredicted the Th(IV) solubilities measured by Rai et al. (1995, Figure 5) at low NaOH concentrations and mostly underpredicted those at high concentrations.

Comparisons of the Th(IV) solubilities measured by Altmaier et al. (2005) in solutions with $I \geq 3$ M with those predicted by FMT (this analysis) show that FMT overpredicted all of these solubilities.

FMT underpredicted all of the Th(IV) solubilities measured by Altmaier et al. (2006) in solutions with $I \geq 3$ M.

Finally, the types and concentrations of colloids reported by Altmaier et al. (2004) are not relevant to the WIPP. Therefore, we concluded that these colloids will not form in the WIPP and did not attempt to assess their effects on PA.

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From: Brush, Laurence H
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Yongliang,

I hereby authorize you to sign for me as a review requester on any of the Document Review and Comment Forms, NP 6-1-1, if necessary; and as an author on my approval line on page 1 of our analysis report "Responses to Three EPA Comments Pertaining to Comparisons of Measured and Predicted Dissolved and Colloidal Th(IV) and Am(III) Concentrations" while I am out of my Carlsbad office in May 2010.

Shannon is putting in the Table of Contents, List of Figures, and List of Tables; and Christi is doing the technical review. When Shannon is done, please have her send it back to me so that I can make the changes that Christi requests. Then I will send it back to you so that you can print it out, sign for you and me, obtain all of the other signatures required for approval, and have Shannon put it into the SNL/WIPP Records Center. Please check the Table of Contents, List of Figures, and List of Tables to make sure that the page numbers are still correct. Sometimes these have gotten scrambled when I have e-mailed this file. If you don't know how to get Word to correct the numbers, please ask Shannon.

Thank you very much in advance!

Larry Brush